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ALTHOUGH the feldspars, the most important group of rock-making minerals, have long been the subject of scientific study and are perhaps better known than any other mineral group, much about them is as yet unknown. The discovery of Köhler, in 1941, that the optical properties of the plagioclase feldspars vary with the temperature of formation has led to a revival of interest in the study of feldspars.

The use of X-rays and the study of silicate systems in the laboratory have opened up new avenues of thought. In this issue of the *Journal* we present a group of papers which embody such studies of the feldspars and the silicate systems related to them.

Among the papers to appear in early issues of the *Journal* are:

"Loess Formations of the Mississippi Valley." By MORRIS M. LEIGHTON and H. B. WILLMAN

"Authigenesis in West Virginia Sandstones." By MILTON T. HEALD

"Pediments and Pediment-forming Processes near House Rock, Arizona." By VICTOR C. MILLER

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September 1950

THE SYSTEM $NaAlSi_3O_8$-$KAlSi_3O_8$-H_2O[1]

N. L. BOWEN AND O. F. TUTTLE
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ABSTRACT

In the presence of water vapor under pressure, soda feldspar and potash feldspar crystallize readily from their glasses. At high temperatures they appear to form a complete series of solid solutions showing continuous variation of lattice-spacing as measured by X-rays. Studies of the dry melts had indicated that the solid solutions are of the type having a minimum-melting mixture. This conclusion is reinforced upon investigation in the presence of water. At a pressure of 1,000 kg/cm² of water vapor the minimum lies at the composition 70 per cent $NaAlSi_3O_8$, and the temperature is 843° C. This temperature is 220° below that observed in dry melts, the lowering being induced by the presence of some 7.5 per cent H_2O in the liquid phase or melt. Increase of pressure to 2,000 kg/cm² produces a further lowering of only about one-third as much. When crystallized at still lower temperatures, the complete solid-solution relation no longer obtains, and two feldspars form side by side. Homogeneous synthetic feldspars formed at high temperatures unmix when held at lower temperatures, in confirmation of the accepted theory of most natural perthites. The composition of both members in natural cryptoperthites can be determined by means of X-ray data established for our synthetic feldspars of known composition.

INTRODUCTION

As the most important group of rock-forming minerals, the feldspars have long been the subject of scientific investigation, and perhaps more is known of them than of any other mineral group; yet much remains to be learned, and their study is still vigorously pursued. Minor constituents aside, the feldspars may conveniently be regarded as solid solutions of three limiting compounds, $NaAlSi_3O_8$, $KAlSi_3O_8$, and $CaAl_2Si_2O_8$, which we may call, simply, soda feldspar, potash feldspar, and lime feldspar, respectively. One of the questions still only partly solved is the extent of solid solution between these components and the manner in which it is affected by conditions prevailing during and after their formation. Another question pertains to their melting behavior. In brief, we need to know their liquidus, solidus, and subsolidus relations as an aid to interpretation of the genesis of natural rocks.

Soda feldspar and potash feldspar and their solid solutions are grouped under the designation alkali feldspars and are thus set off from the other feldspar series, the soda-lime feldspars or plagioclases—a convenient, if oversimplified, classification. There are igneous rocks composed largely or almost entirely of alkali feldspars. The deep-seated, intrusive varieties are called syenites, and their surface or lava equivalents are called trachytes.

Syenites and trachytes do not loom large in the igneous economy, but with an excess of silica they pass into granites, which are the most abundant rocks of the

[1] Manuscript received April 10, 1950. Presented at the November, 1949, Annual Meeting of the Geological Society of America.

earth's crust; and, when silica is deficient, they pass into those interesting, if not very abundant, rocks, the nepheline syenites, which find their surface equivalents in the phonolites, just as the granites are represented as lavas by rhyolites and obsidians. A study of the equilibrium relations of the alkali feldspars is thus a necessary preliminary to an understanding of these chemically related groups of igneous rocks. The evidence is strong that the liquids or melts from which many, if not all, of these rocks have consolidated contained significant quantities of water dissolved in them under pressure. For a proper understanding of the rocks it is therefore necessary to examine the melting equilibrium relations of the alkali feldspars under measured pressures of water vapor. Though the experimental techniques are more complicated than those applicable to dry melts, an enormous advantage accrues in that attainment of equilibrium is facilitated not only at liquidus temperatures but also at lower temperatures, which are particularly important in the present case.

PREVIOUS INVESTIGATIONS

From studies of natural alkali feldspars and their occurrences, some very astute conclusions have been reached on the questions of their equilibrium relations, but a review of such work is beyond the scope of this paper. A number of experimental studies on natural feldspars designed to throw light on their thermal relations have also been made. We shall not attempt to review this work either, but references to specific aspects of it will be made later.

Studies of synthetic alkali feldspars have also been carried out, but extraordinary difficulty is encountered in obtaining crystallization of these compositions in the dry way. Some progress has been made, however, and a preliminary paper presenting results was published by Schairer and Bowen (1935). Schairer has since carried the work further, and his results are given in a companion paper herewith.

Moreover, the melting relations of soda feldspar and potash feldspar individually, in the presence of water at high pressures, have been examined by Goranson (1938). Our task was to extend these studies to mixtures of the feldspars.

EXPERIMENTAL METHODS

The apparatus used in our investigation was principally the hydrothermal quenching apparatus of Tuttle (1948). The details of its use in a routine investigation have already been presented (Bowen and Tuttle, 1949). Suffice it to say here that the charge was of about 10 mg. in the form of powder and was placed in a small platinum crucible, which was covered with another tightly fitting platinum crucible, and these were then placed in a somewhat larger crucible, the space between then being filled with powder identical with the charge in composition. This surrounding buffer had been found necessary in the earlier work in order to avoid change of composition of the charge through transfer of material in the vapor phase by convective circulation.

The whole was then exposed to a measured and controlled temperature and pressure of water vapor in the manner described in the reference given. On occasion, other forms of apparatus, designed by Tuttle (1949), were also used.

In nearly all the experiments or "runs," the initial material was a homogeneous glass, containing the two feldspar "molecules" in the desired proportions, which had been prepared by

Schairer for the work with the dry melts. The preparation of these glasses is an extraordinarily difficult and tedious process, and their availability from the previous work was of inestimable value in the present studies.

Further details of procedure designed to reach specific ends will be given as occasion requires.

APPARENT UNBROKEN SERIES OF SOLID SOLUTIONS AT HIGH TEMPERATURES

In the dry way, glasses of the composition of the alkali feldspars refuse to yield a completely crystalline product at any temperature, even if the duration of crystallization is measured in years. Without completely crystalline material, temperatures of beginning of melting cannot be determined. Partial crystallization can, however, be realized, with sufficient patience and persistence, and on the partially crystallized material temperatures of completion of melting (liquidus) can be determined. The results are given in the companion paper of Schairer. The best interpretation of the results is that soda and potash feldspars form a complete series of solid solutions, of the type with a minimum-melting composition, the diagram being complicated toward the potash-rich side by the incongruent relations resulting from the separation of leucite. However, three compositions give an identical liquidus temperature, within the error of measurement, and the possibility remains that we are dealing, not with a very flat minimum, but with a eutectic, which would necessitate a hiatus in the solid-solution series. Further evidence is desirable, and, indeed, conclusive evidence is found through studies in the presence of water.

When glasses of the system are crystallized at 900° C. under a pressure of water vapor of 300 kg/cm², complete crystallization of all compositions is readily attained. It is now possible to measure various physical properties on these crystalline products of known composition and to determine the change of properties with composition.

The X-ray powder charts obtained by means of the Phillips X-ray spectrometer show a continuous change with composition. A series of these is shown in figure 1. The continuity is even more convincingly shown in a series made up of more closely spaced compositions; but for a quantitative test we selected for accurate measurement with the aid of the counting circuit one of the maxima or "peaks" on the chart which has a position corresponding with a 2θ value of about 21° for potash feldspar and about 22° for soda feldspar (for CuKα radiation) and measured its position for thirteen intermediate compositions. The absolute position of the peak for each composition was determined by mixing with the feldspar powder some olivine powder, which has a peak corresponding with a 2θ value of about 23°, and determining the difference between this olivine peak and the feldspar peak under investigation. The olivine peak was, in turn, calibrated against a quartz peak having a 2θ value of about 20°.85, which is somewhat too close to the values for the potash-rich feldspars to permit the use of quartz as a direct reference material for all the feldspars, although it could be used directly for all but those richest in potash. The reflection angles were transformed into spacings of the $(\bar{2}01)$ planes in the feldspar crystals,[2] which are, in accord with the above considerations, referred to the $(10\bar{1}0)$ spacing of quartz taken as 4.2549 Å. The values are plotted

[2] The plane was indexed by using the results of Taylor (1933) on natural sanidine.

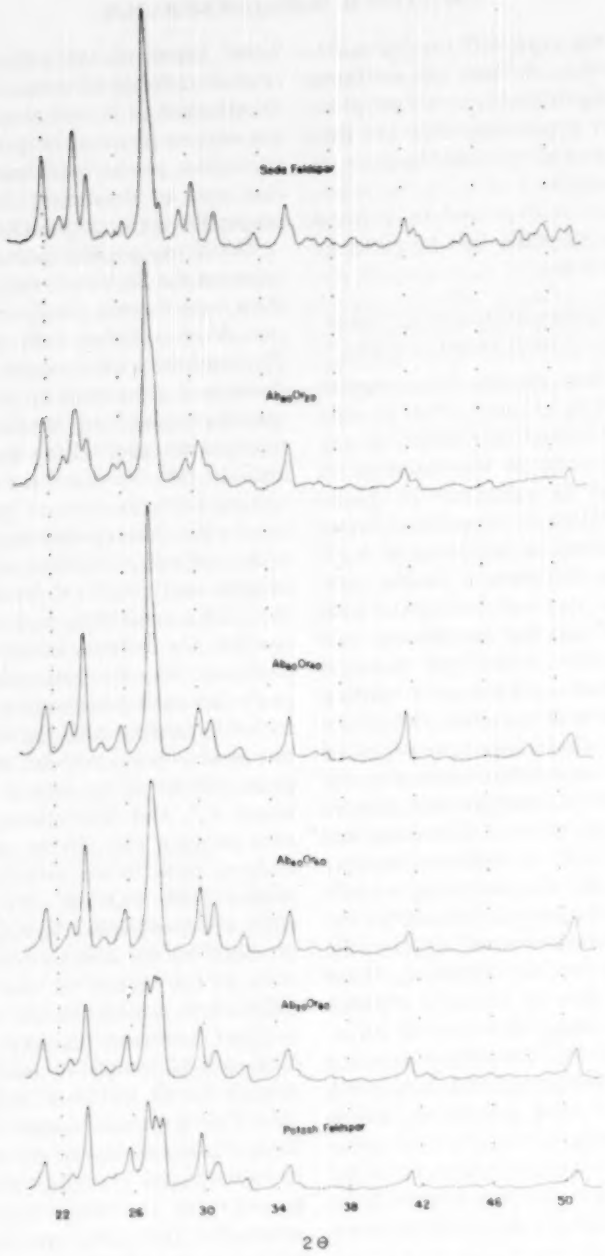

FIG. 1.—X-ray diffraction charts of alkali feldspars crystallized at 900° C. and 500 kg/cm² pressure of H_2O.

in figure 2 against the composition of the feldspar in weight per cent. They exhibit a linear relation. Because the change of composition is the result of substitution of K^+ for Na^+, which is the same as substitution of $KAlSi_3O_8$ for $NaAlSi_3O_8$, for any consideration of fundamentals, the values should presumably be referred to composition expressed in mol per cent. So plotted, there is appreciable departure from the linear relation. However, all the compositions were made up in weight per cent, and a linear diagram is so convenient that we retain it in that form. The continuous change of spacing from one end-member to the other seems to be satisfactory evidence of complete solid solution. The error of measurement of 2θ for any composition is somewhat less than 2 per cent of the total change of 2θ. The possibility of the existence of two curves of different slope, with a hiatus in which two feldspars exist, is not suggested by these results. To be sure, the peaks for two feldspars close together in composition are not resolved, but the resultant peak is broadened, and no evidence of this effect was detected in any composition.

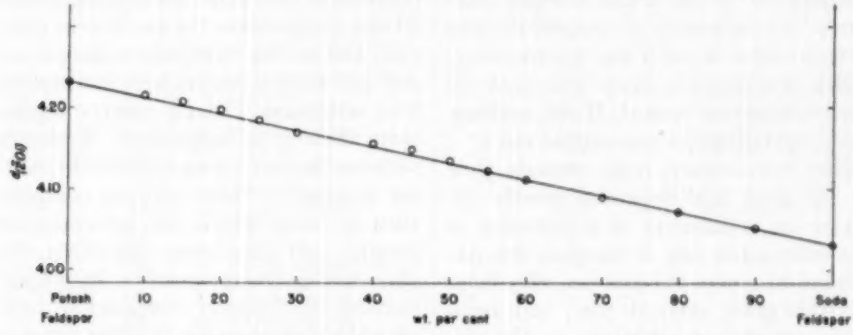

FIG. 2.—Spacing of $(\bar{2}01)$ planes in a series of alkali feldspars crystallized at 900° C. and 300 kg/cm² pressure of H_2O.

It will be recalled that the foregoing apparently complete series of solid solutions was obtained by crystallizing at 900°. At low temperatures very different results are obtained; but, before describing these, we shall first consider further evidence indicating the existence of a complete series of solid solutions at high temperatures. This comes from the determination of the melting relations in the presence of water vapor at high pressures. As has been stated already, only the liquidus curve can be determined in dry melts, and some doubt is left as to the exact relations between the feldspars, but the determination of both solidus and liquidus, which is readily accomplished under pressure of water vapor, leaves little room for doubt. The foregoing X-ray data, obtained on crystals of known composition, were of great service in establishing the solidus curves at different pressures, as we shall see.

THE EQUILIBRIUM DIAGRAMS AT VARIOUS PRESSURES OF WATER VAPOR

We have found it convenient to investigate equilibrium at isobars, that is, to work at constant pressure with various compositions and then to repeat at other constant pressures. Such procedure yields diagrams that are so simply related to the "binary" diagram for the dry melts that

they are readily understood. Indeed, the diagram for the dry melts is itself the isobaric diagram for the pressure of water vapor in the atmosphere, which is, for practical purposes, zero.

In an individual run with any mixture the initial material was always a glass of known composition. By experience it was found necessary to grind the glass very fine and then to crystallize at a temperature of 700° and 1,000 kg/cm² water pressure for 30 minutes to 1 hour. This procedure gave a very fine-grained, completely crystalline product of uniform composition for all alkali feldspar mixtures. The uniformity of composition was determined with the X-ray spectrometer, which gave only one sharp ($\bar{2}01$) peak for each mixture so treated. If this preliminary crystallization was carried out at a higher temperature, large crystals grew in the glass, and their size greatly delayed the attainment of equilibrium in the subsequent run. If the glass was not ground to a very fine powder, very large crystals grew, even at 700°, and again there was the same difficulty in the subsequent run. If crystallization was carried out below 700°, two feldspars resulted, as indicated by the presence of two peaks for ($\bar{2}01$). But, by following the procedure indicated and obtaining a very fine-grained product consisting of one feldspar of uniform composition, it was possible with this material to determine the temperature of beginning of melting, the temperature of completion of melting, and other relations to be mentioned. This was accomplished by holding the charge thus pretreated at constant temperature and pressure for the desired period, quenching it, and then examining the product with the petrographic microscope and the X-ray spectrometer. For all runs the charge was approximately 10 mg. in weight. The results of the runs are set forth in table 1 and are shown diagrammatically in figure 3.

The figure speaks for itself, and little comment is needed. However, some further details of procedure may now be given that are more readily appreciated with the aid of the diagrams. The liquidus curve for each pressure is, of course, determined by finding for each feldspar composition the temperature at which the last crystals disappear. The solidus curve is determined by finding the temperature at which the first liquid appears. At one composition the two curves coincide, and at this composition both liquidus and solidus curves have a temperature minimum. For all other compositions there is a temperature difference between the two curves which is the melting interval for that feldspar composition. A charge held in this interval gives crystals and glass upon quenching, the glass having the composition (in proportions of the feldspar components) indicated by a point on the liquidus curve at the temperature of the run and the crystals having the composition given by the point on the solidus curve at the same temperature. This fact enables the fixing of the solidus curve much more accurately than it could be fixed merely by observing the temperature at which liquid first appears, for it is difficult to detect a small proportion of glass in a fine crystalline aggregate. It is necessary only to hold a charge at a measured temperature in the melting interval, quench it, and then determine the composition of the crystals by means of the X-rays, using the straight-line relation of figure 2. A point on the solidus for the temperature of the run is thus determined. This, which may be termed the "composition method," is much more accurate than

TABLE 1

Results of Quenching Experiments on Alkali Feldspar Compositions at Various Temperatures and Pressures of Water Vapor

Liquidus				Solidus			
Comp. (Wt. Per Cent) Ab	Temp.	Time (Hours)	Result	Comp. (Wt. Per Cent) Ab	Temp.	Time (Hours)	Result
	P_{H_2O} = 500 Kg/Cm²						
60	921	48	All glass	50	920	72	Glass and xls (Ab_{41})
70	900	24	All xline	60	910	72	Glass and xls (Ab_{54})
80	900	70	All xline	65	902	72	Glass and xls (Ab_{61})
80	930	48	Glass and rare xls	70	925	36	Glass and xls (Ab_{72})
				80	930	20	Glass and xls (Ab_{80})
	P_{H_2O} = 1,000 Kg/Cm²						
100	900	72	Glass and trace xls	80	862	20	Glass and xls (Ab_{85})
100	880	20	All xls	70	844	20	Glass and xls (Ab_{70})
85	885	20	All glass	60	855	72	Glass and xls (Ab_{47})
85	875	20	Glass and xls	50	854	20	Glass and xls (Ab_{44})
85	850	20	All xls	50	880	72	Glass and xls (Ab_{44})
80	862	20	Glass and xls	30	885	70	Glass and xls (Ab_{16})
70	855	72	All glass	15	930	20	Glass and xls (Ab_8)
70	844	20	Glass and xls	15	910	20	Glass and xls (Ab_9)
70	840	72	All xls				
60	880	7	All glass				
60	855	70	Glass and rare xls				
50	900	20	All glass				
50	880	72	Glass and rare xls				
50	854	20	Glass and xls				
40	920	12	All glass				
40	900	12	Glass and xls				
30	965	24	Glass and rare xls				
30	947	12	Glass and xls				
30	930	20	Glass and xls				
30	900	20	Glass and xls				
25	1,000	2	Glass and leucite xls				
25	980	2	Glass and leucite xls				
15	1,000	2	Glass and leucite xls				
15	980	3	Glass, leucite, and feldspar				
15	930	20	Glass and feldspar				
0	1,000	4	Glass and leucite				
0	980	4	All xls (feldspar)				
	P_{H_2O} = 2,000 Kg/Cm²						
80	800	20	Glass and rare xls	80	800	20	Glass and xls (Ab_{88})
70	790	12	All glass	70	767	72	All xls
60	805	18	Glass and xls	70	780	20	Glass and xls (Ab_{70})
60	820	18	All glass	60	770	20	All xls
50	835	16	Glass and xls	60	795	70	Glass and xls (Ab_{53})
50	850	17	All glass	60	778	20	Glass and xls (Ab_{57})
40	860	6	Glass and xls	50	825	20	Glass and xls (Ab_{50})
40	875	4	All glass	50	832	20	Glass and xls (Ab_{51})

Table 1—*Continued*

SOLVUS (P_{H_2O} = 1,000 Kg/Cm²)

Comp. (Wt. Per Cent) Ab	Temp.	Time (Hours)	Result
	All Charges Initially Glass		
55	670	170	Sharp peak (1 feldspar)
55	660	170	Sharp peak (1 feldspar)
55	650	170	Broad peak (2 feldspars)
55	640	170	2 equal peaks (2 feldspars)
60	600	170	2 feldspars (Ab_{32} and Ab_{80})
60	500	170	2 feldspars (Ab_{25} and Ab_{93})
	Charge Initially Crystallized as Single Feldspar		
55	530	170	2 feldspars (Ab_{36} and Ab_{72})

the temperature method of determining the solidus.

The minimum on the solidus (and liquidus) curve is determined by applying the same principle. It is clear from a glance at the equilibrium diagrams that the lower part of the solidus is so flat that only a very rough approximation to the composition of the minimum could be reached by the temperature method. On the other hand, the minimum can be located very accurately by the composition method. If the composition chosen for investigation lies to the right of the minimum, the crystals move in composition to the right as melting proceeds; if the composition lies to the left of the minimum, they move to the left; and if it lies at the minimum and is heated for a period of time which is insufficiently long for complete melting, the composition of the crystals remains as it was initially. Actually, it was found that crystals with 70 per cent soda feldspar melt without measurable change of composition at both 1,000 and 2,000 kg/cm² pressure of H_2O; but in all other compositions the crystals change in composition in one direction or the other, according to composition, as indicated above. The composition of the minimum is therefore placed at this value for both these pressures. The actual minima could depart from one another by about 2 per cent without our being able to detect the departure with certainty, but not by any significantly greater value.

Now in the diagram for the dry melts there are three compositions, 60, 65, and 70 per cent soda feldspar, all of which melt at 1,063°, within the limits of error of measurement, and it was natural, therefore, to place the minimum at the intermediate value, 65 per cent. The difficulties of experimentation with the dry melts are so great and the placing of our minima at both pressures at 70 per cent has so small a probable error that we at first supposed that the minimum probably should be at the same value in the dry melts also. In order to test this possibility, we made a run with the 70 per cent composition at a low pressure of water vapor (500 kg/cm²). It was found

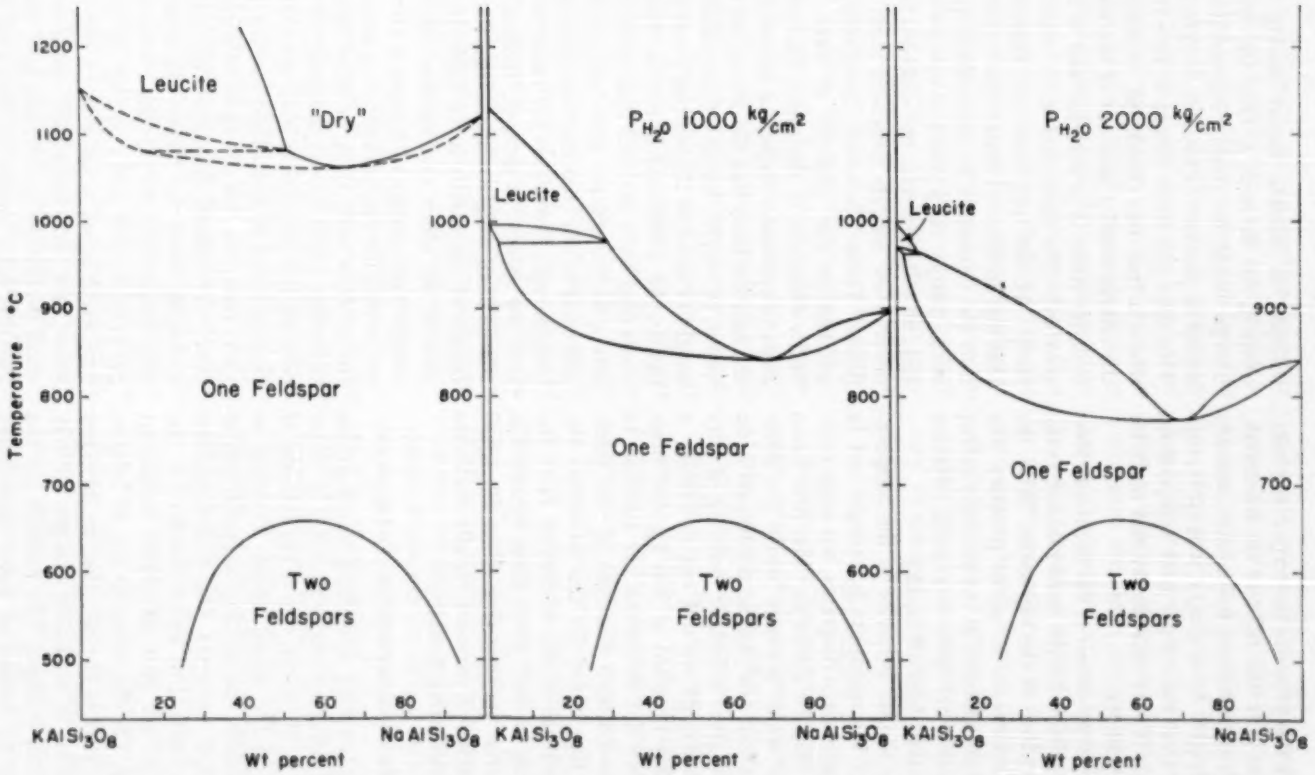

FIG. 3.—Isobaric equilibrium diagrams for the alkali feldspars in dry melts and at 1,000 kg/cm² and 2,000 kg/cm² pressure of H_2O

that at this pressure the 70 per cent composition is to the right of the minimum, for the crystals now move unmistakably to the right. Accordingly, it must be supposed that the 65 per cent composition actually is very close to the minimum for the dry melts.

The composition of the minimum thus behaves in much the same manner as the temperature of the minimum. With the first increments of water pressure, the minimum changes in composition rather markedly, but with increasing pressure the change falls off rapidly.

The similar behavior of the temperature of the minimum is brought out in the equilibrium diagrams, but some comment may be desirable. The first 1,000 kg/cm² water pressure lowers the minimum about 220°, but an additional 1,000 kg/cm² gives a further lowering of only 73°. Although we did not determine a complete diagram at 500 kg/cm² pressure, we did determine (in addition to the composition relations at the minimum just noted for this pressure) the temperature of the minimum. Here the lowering is 160°. From these figures it is clear that the lowering effect for equal increments of pressure rapidly decreases with increasing pressure.

WATER CONTENT OF THE LIQUID PHASE

The lowering of melting temperatures of the feldspars under pressure of water vapor is, of course, due to the solution of water in the liquid phase. Pressure as such raises the melting temperatures of the alkali feldspars. Our determinations of the content of water dissolved in the glass do not give altogether consistent results. The discrepancies are, we believe, due in part to the fact that we have not succeeded in obtaining charges entirely free from bubbles. The bubbles are not formed by release of water vapor from solution in silicate liquid during the quench, for in such a case the whole charge would be greatly expanded and would be jammed in the stellite cylinder. This does not occur except in rare runs, where a leak has developed. Ordinarily the charge readily falls out of the metal cylinder when it is inverted and is substantially the same size as it was initially. We can only conclude, therefore, that the bubbles are water which entered into the interstices of the initially powdered charge. As a result of surface tension or other effects, the bubbles have never been entirely liberated from the liquid. These, therefore, constitute an addition to the quantity of water actually dissolved in the glass. Viewed at room temperature under the microscope, the bubbles have two phases, liquid and vapor, in roughly equal volume, and the weight of water in the bubbles is not negligible. An additional source of error is connected with the fact that water dissolves in the silicate glass very slowly, and some of our runs may not have been long enough. Error from this cause would give low values. In general, because the charge itself weighs only 10 mg., the accuracy of our results is not high. In the case of the albite end-member, we can compare our results with those of Goranson, which themselves cannot be entirely free of error from these or other causes. Goranson (1938, p. 82) obtained 5.8 per cent as the average of two determinations of water in albite liquid at 910° and 1,007 bars; but, on the basis of a calculated curve passed through all his values, including those for higher pressures at this temperature, he accepts a value of 6 per cent. At 900° and 1,000 kg/cm² pressure we obtained a value of 5.6 per cent water, but the charge still contained a small proportion of crystals, and therefore this value should be somewhat low.

At 910° and 1,000 kg/cm² pressure we obtained a value of 6.9 per cent, which is probably high. The agreement between our results and Goranson's is, therefore, satisfactory, if one is content with that degree of approximation. The accurate determination of the amount of water dissolved in these silicate melts would be a major research, which must await a future time. We have made only a few determinations, and of these we present only those just mentioned and those obtained on the minimum-melting mixture (70 per cent $NaAlSi_3O_8$) at the two pressures 1,000 and 2,000 kg/cm² and at a temperature only a few degrees above its liquidus. The best values appear to be for p = 1,000 kg/cm², t = 854°, water content = 7.5 per cent; and p = 2,000 kg/cm², t = 785°, water content = 9.0 per cent. For such intermediate compositions there are no determinations by others available for comparison, but the values are consistent with those obtained by Goranson on $NaAlSi_3O_8$ and $KAlSi_3O_8$ separately, when allowance is made for the fact that, at the liquidus at any pressure, the amount of water dissolved in the liquid should be higher at the composition of the minimum-melting mixture than in the end-components.

THE LEUCITE FIELD

Leucite is the primary phase in the composition $KAlSi_3O_8$ and is the stable phase in contact with liquid through an interval of upward of 400°. It is not surprising, therefore, that the field of leucite extends far out on the feldspar join. Actually, it extends to 51 per cent albite in the dry melts, as is shown in figure 3. Goranson found that, in $KAlSi_3O_8$, water greatly diminishes the temperature interval in which leucite forms. At 1,000 kg/cm² pressure of water the interval is reduced to about 135°. In correspondence with this, we find that the leucite field extends only to 29 per cent albite at 1,000 kg/cm². At 2,000 kg/cm²-pressure of water, the leucite field is reduced to very small dimensions, so that nearly the whole feldspar diagram shows simple congruent relations. According to Goranson's determination, at or near 2,500 kg/cm² pressure of water, the leucite field is suppressed entirely in $KAlSi_3O_8$. At this pressure of water, therefore, the whole feldspar diagram should become simple, but we have not thought it necessary to confirm this, since the trend is clear.

THE SOLVUS

As has been indicated on an earlier page, when alkali feldspar glasses are crystallized at low temperatures with water vapor under pressure to facilitate crystallization, instead of a single homogeneous feldspar, two feldspars are formed. This fact is established from the observation that two maxima or peaks of X-ray reflection from ($\bar{2}01$) are shown; and by measuring the exact position of these peaks by means of the counting circuit and referring them to the linear relation of figure 2, the composition of the two feldspars in equilibrium at any temperature can be determined. It follows that a homogeneous feldspar formed at high temperatures should unmix at lower temperatures, a relation which was long ago deduced from natural materials (perthites), but now we have a means of establishing the position of the unmixing curve or solvus.

The solvus is shown in each of the diagrams of figure 3. Unlike the solidus and liquidus curves, the solvus does not change its position measurably with change of pressure of water vapor because water does not participate in the equilibria involved at the solvus, whereas

it does participate in the solidus-liquidus equilibriums by entering into the composition of the liquid phase. Pressure as such has, of course, some effect on the solvus, but the magnitude of the effect is relatively very small because it depends on the volume change attending mutual solution (solid solution) of the two feldspars and this volume change is small.

The three diagrams of figure 3 show that, unless some new factor intervenes—for example, critical phenomena—there will be a pressure of water vapor which will lower the solidus-liquidus curves to temperatures at which they will intersect the solvus. At this isobar and for all isobars corresponding to higher pressures, the feldspar diagram will cease to be one showing complete solid solution. Instead, there will form two series of solid solutions with a hiatus and a eutectic or a reaction relation, depending on the manner of encounter of the solidus and solvus. The indications are that it will first be a reaction relation which at still higher pressures gives way to a eutectic relation.

The solvus has its maximum temperature at 660° ± 10°, at a composition close to 55 per cent soda feldspar. Thus 660° is the minimum temperature of stable existence of a feldspar of that composition, and each composition of feldspar has its own minimum temperature of stable existence at which it will, if opportunity for equilibrium prevails, begin to break up or unmix into two feldspars. These depart more and more widely from each other in composition as the temperature falls. Of course, when crystallized at 700° or higher, any feldspar can be cooled rapidly to room temperature and will remain a single homogeneous feldspar—indeed, that is the method whereby we determined that a complete series exists at 700° and higher. But if any feldspar is held at a temperature where it should form two feldspars, as indicated by the solvus, unmixing will occur in a period of a few days if water vapor under pressure is present to facilitate mobility. This behavior confirms the results obtained by direct crystallization of glass at various temperatures, which is the method whereby the solvus was determined. The presence of two feldspars in the product obtained by thus inducing unmixing from an originally homogeneous feldspar cannot, to be sure, be observed with the petrographic microscope. The intergrowth is submicroscopic and is revealed only by the X-rays.

An intergrowth of two feldspars thus obtained by unmixing from a single feldspar can be rehomogenized by heating for about 24 hours at 700° even in the absence of water vapor, and at higher temperatures homogenization is accomplished in shorter times as, for example, in 1 hour at 950°.

The mixing and unmixing of alkali feldspars is thus readily reversible, as is shown by the behavior of these submicroscopic intergrowths, and their behavior is altogether in accord with the determined solvus. There is, however, one feature of the solvus as thus determined in the laboratory that must now be emphasized. It is the possibility that at least some part of our solvus may pertain to metastable equilibrium. All the feldspars obtained synthetically in these studies are high-temperature varieties, and the solvus is the unmixing curve of these high-temperature feldspars. At 500°, for example, our solvus shows that the soda-rich feldspar has a composition approaching that of pure albite. It is, however, possible that the soda-rich feldspar stable at 500° is the low-temperature variety and not the high-temperature form which we obtained. It is a common experience with silicates to ob-

tain a high-temperature form metastably at low temperatures. The only indication that we have as to the inversion temperature of albite was obtained by finding the lowest temperature at which natural low albite could be transformed to high albite with the aid of a flux, a procedure which gave 700°. From this evidence it can only be said that the inversion is not higher than 700° but that it could be lower. If it is as low as 500°, then our solvus, as determined down to 500°, represents stable equilibrium. If the inversion is really as high as 700°, then the lower-temperature part of the soda-rich limb of the solvus may refer to metastable equilibrium.

However, even if the inversion of albite is as high as 700°, it is not necessary that any part of the soda-rich limb of our solvus should be metastable. It is highly probable that the addition of potash feldspar to albite will lower its inversion temperature, for, almost certainly, potash feldspar will be less soluble at any temperature in low albite than in high albite. A quite probable form of equilibrium diagram would therefore be one such as that given in figure 4. In this we have retained all our determined solvus and yet placed the albite inversion at 700°. There are also some complications in connection with the potash-rich limb of our solvus, as we shall see when we turn to natural feldspars.

BEHAVIOR OF NATURAL ALKALI FELDSPARS

It has long been known that, in rocks of appropriate composition which are believed on geologic grounds to have crystallized at high temperatures, a single feldspar with high content of both alkalis is normally found, whereas in rocks believed to have formed at low temperatures a soda-rich feldspar and a potash-rich feldspar are found side by side. These observations, together with other supporting evidence to be mentioned, have led to the conclusion that the alkali feldspars form a nearly, if not entirely, complete series of solid solutions at high temperatures but have only limited miscibility in the crystal state at low temperatures, a conclusion now demonstrated to be true by our investigation of synthetic feldspars.

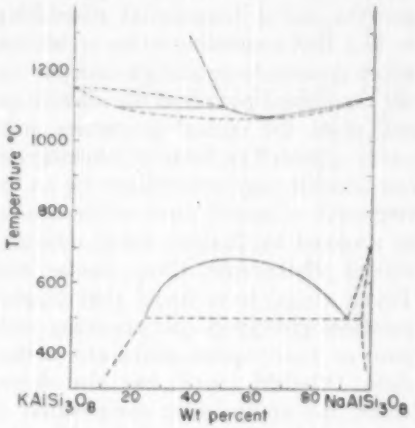

FIG. 4.—A completion of the sodic end of the alkali-feldspar diagram, showing the probable relations of high and low albite.

The existence in nature of perthitic, microperthitic, and cryptoperthitic intergrowths of the two feldspars supported the early conclusion and indicated also that even when a homogeneous feldspar is first formed it may unmix in the solid state at lower temperatures, giving an intergrowth of the two feldspars. X-ray studies have served to reveal the fact that two feldspars exist in the "crypto" varieties of these intergrown feldspars, such as the moonstones (Kozu and Endo, 1921); and by heat treatment some investigators (Kozu and Endo, 1921; Spencer, 1930, 1937) have

shown that the feldspars will again form a homogeneous solid solution.

These conditions and changes of condition are all readily produced in the synthetic feldspars, as we have seen, and we have been able, in addition, to determine by the X-rays the composition of each of the feldspars in the synthetic intergrowths. This, too, we have been enabled to do with the natural crypto-intergrowths, and it is somewhat astonishing to find that a sanidine or an orthoclase which appears to be a single feldspar under the highest powers of the microscope and gives the optical properties ordinarily regarded as those of a homogeneous feldspar may nevertheless be an intergrowth of nearly pure end-members, as revealed by X-rays. Using rotating-crystal photographs, Chao, Smare, and Taylor (1939) have found that cryptoperthites give spots corresponding with pure or nearly pure albite and orthoclase. Oftedahl (1948) has also determined the approximate composition of the two feldspars in cryptoperthites by the rotating-crystal and Weissenberg methods.

We must now emphasize, however, that, when examined further, the actual relations in the natural feldspars are considerably more complex than those in our synthetic analogues. These latter, throughout all the changes to which we have subjected them, retain their characteristics as high-temperature feldspars. The only change that a synthetic intergrowth suffers on heating is homogenization, which is presumably accomplished merely by mobilization and diffusion of the cations.

The natural intergrowths, on the other hand, are, in many instances, low-temperature feldspars. When these feldspars are heated, the cations are readily mobilized, and they give, within certain limits of composition, a homogeneous feldspar after a short period of heating; but the feldspar so produced is quite different from one grown at a high temperature or produced by re-homogenizing an intergrowth of two high feldspars. The optical relations of these natural feldspars heated only for the short period adequate for homogenization are those characteristic of low feldspar. Spencer (1930, 1937) has made an excellent detailed study of these relations, and our results on natural feldspars are in substantial confirmation of his. Only after prolonged heating at temperatures but little below its melting temperature can a homogenized natural feldspar of the low-temperature variety be transformed into a feldspar having the optical properties of a high-temperature feldspar. This change is presumably accomplished by mobilization of other atoms (ions), namely, Si and Al, giving a disordered condition, their mobilization being much less readily induced than that of the cations. Our X-ray studies enable us to add a little to the information obtained by Spencer, but the details must await a later communication.

GENERAL CONSIDERATIONS ON EQUILIBRIUM

Goranson (1938) has given us a very complete study of equilibrium between soda-feldspar crystals and liquid (melt) under various pressures of water vapor up to nearly 3,000 kg/cm². In figure 5 is given a theoretical completion of the *PTX* diagram (in the three projections *PT*, *PX*, and *TX*) for water-$NaAlSi_3O_8$, which embodies his determinations. The figure shows the conventional critical end-points *P* and *Q*, whose exact locations are, of course, quite unknown; but the plain trend of Goranson's results indicates that, no matter how high the

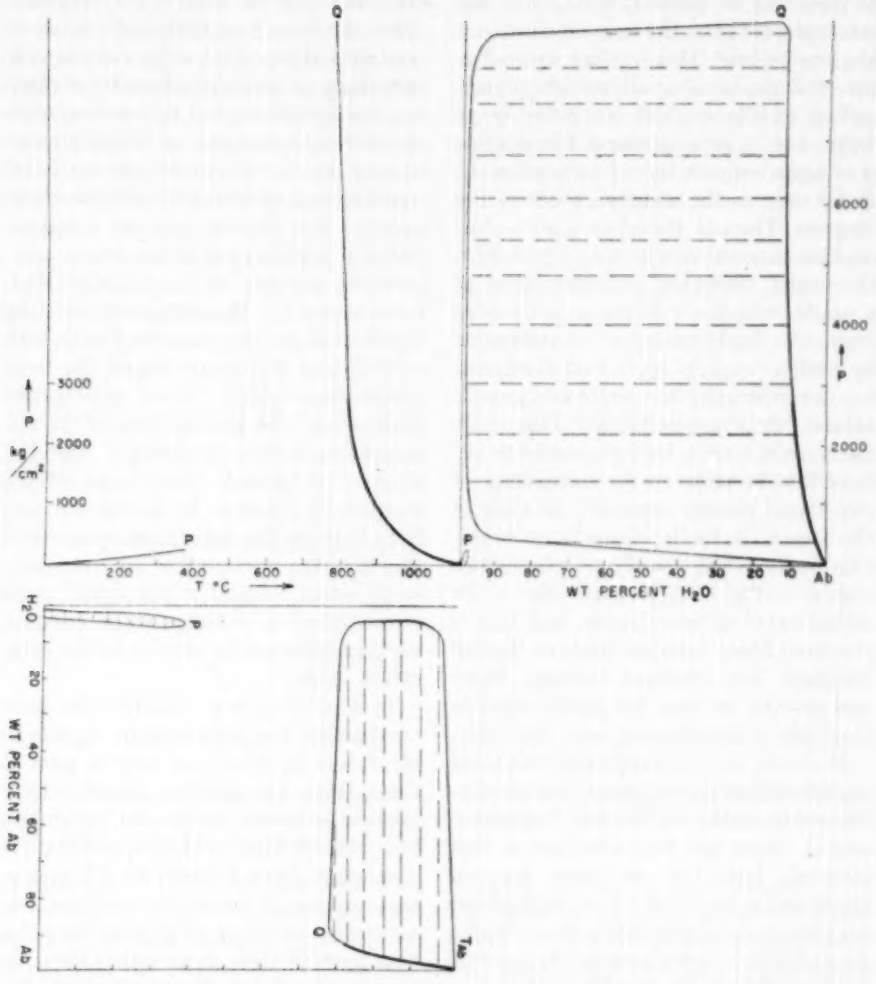

FIG. 5.—Theoretical completion of the PT, PX, and TX diagrams for $NaAlSi_3O_8$-H_2O. Only the heavy portions of curves are based on determinations (Goranson). P and Q are the first and second critical endpoints. In the TX projection, $T_{Ab}-Q$ (clockwise) gives the composition of all liquids and $T_{Ab}-Q$ (anticlockwise) gives the composition of all vapors in equilibrium with albite crystals. In the PX projection Ab$-Q$ (anticlockwise) gives the composition of all liquids and Ab$-Q$ (clockwise) gives the composition of all vapors in equilibrium with albite crystals.

pressure, the amount of water that can be dissolved in $NaAlSi_3O_8$ liquid is not much greater than the amount dissolved at 3,000 kg/cm². This fact is indicated in the *PX* projection, albite-rich liquids ending at the critical end-point *Q* at some 12 or 13 per cent water. Liquids appear again only at the critical end-point *P*, far over on the water-rich side of the diagram. There is, therefore, a wide composition interval in which no liquid solutions exist. Goranson's demonstration of a similar reluctance of water to dissolve in granitic liquid has led to the statement by Smith (1948, p. 536) that Goranson has demonstrated that water and granite exhibit "fluid immiscibility." This statement could readily lead the reader to believe that he refers to the coexistence of two liquid phases, especially in view of the sequel. Actually, there is no condition under which two liquid phases can coexist in the *PTX* diagram (fig. 5) for albite-water as here drawn, and this is the most likely interpretation of the full diagram. The relations between water and granite, at least the granite used in Goranson's experiments, are the same.

It should be noted, however, that these considerations do not mean that no solutions exist in the *PT* interval between *P* and *Q*. There are fluid solutions in that interval; but, because there are not liquid and vapor both, not enough phases are present for univariancy. Accordingly, no univariant curve crosses the interval between *P* and *Q* in the *PT* diagram.

With this interpretation of $NaAlSi_3O_8$-water equilibria before us, we may pass to a consideration of our present system. For convenience we may, for the moment, neglect the incongruent melting of $KAlSi_3O_8$ and assume that it would give a diagram with water analogous to that for $NaAlSi_3O_8$. Then in the ternary system we would have the relations shown in the two diagrams of figure 6, the one a *PX* and the other a *TX* diagram.

In discussing equilibrium between crystals and liquids in a dry system, it is customary to speak of a liquidus surface or a fusion surface, but in a system with water these terms are not entirely satisfactory. In such a system interest is focused upon a very special fusion surface, namely, that representing the composition of liquids in equilibrium at any temperature, not only with crystals but with water vapor. For the surface representing liquids that are thus saturated both with crystals and with water vapor, the term "saturation surface" is an appropriate designation. The general form of the *TX* saturation surface is shown in the *TX* diagram of figure 6. There is an infinite number of liquidus or fusion surfaces lying between the saturation surface and the liquidus surface for an extremely small water content in the liquid, each representing a definite water content, varying from nearly zero up to the saturation value.

In the foregoing remarks we have emphasized the temperature aspects of saturation because it is easy to pass to them from the familiar considerations applicable to dry melts. But saturation has pressure aspects as well, and the *PX* diagram of figure 6 shows the *PX* saturation surface. A knowledge of these two saturation surfaces, or at least the pertinent parts of them, is necessary for a discussion of courses of crystallization. For an understanding of the general relations, reference should be made to the legend of figure 6.

ISOBARS AND ISOTHERMS ON THE SATURATION SURFACES, AND THE COURSES OF CRYSTALLIZATION

The *PX* and the *TX* surfaces are utterly unlike and are not usable in them-

selves, but one can obtain a most useful diagram by projecting them, or that part of them which has been experimentally determined, upon the composition triangle, representing the one by isotherms and the other by isobars. This has been done in figure 7, which enables one to describe completely the crystallization of any liquid within the limits of our determinations. The isobaric sections that are presented in figure 3 and represent the simplest experimental approach to the solution of the problems of the system nevertheless do not serve to describe crystallization completely because the water content of the liquid is not shown in such sections, although it was, of course, the data shown in these sections plus the determination of water in the quenched glasses that enabled us to construct figure 7, showing saturation isobars and isotherms on the composition triangle.

Actually, the "composition triangle" here presented is really only a part of that triangle, and it is constructed in an unusual manner, which requires some description. Although it is customary to represent ternary compositions on an equilateral triangle, it is well known that one can use a triangle of any shape whatever with co-ordinates drawn parallel to its sides. For the present purpose it is convenient to use a triangle in which the water ordinate is greatly exaggerated, and, accordingly, we have constructed an isosceles triangle with an altitude five times that of an equilateral triangle. Water is plotted at its apex, and one of the feldspars at each end of its base. Of this triangle we show only the lower portion in figure 7 because it is the only portion required to represent the experimental results. The proportion of water is read in the ordinary manner with the aid of lines parallel to the base, but, as it is

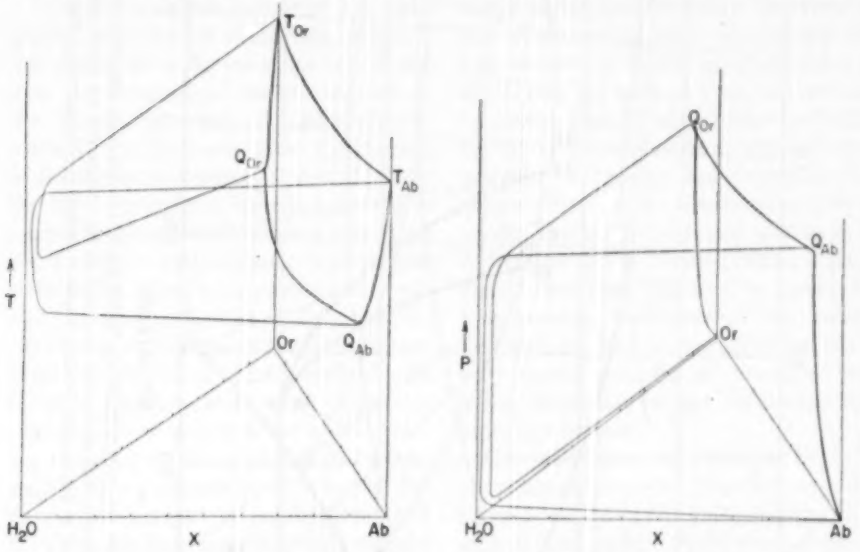

FIG. 6.—Schematic *TX* and *PX* diagrams for the system Ab-Or-H_2O, to aid in the understanding of figure 7. T_{Ab}-T_{Or}-Q_{Or}-Q_{Ab} is the temperature saturation surface of alkali feldspar crystals, and Ab-Or-Q_{Or}-Q_{Ab} is the pressure saturation surface of those crystals.

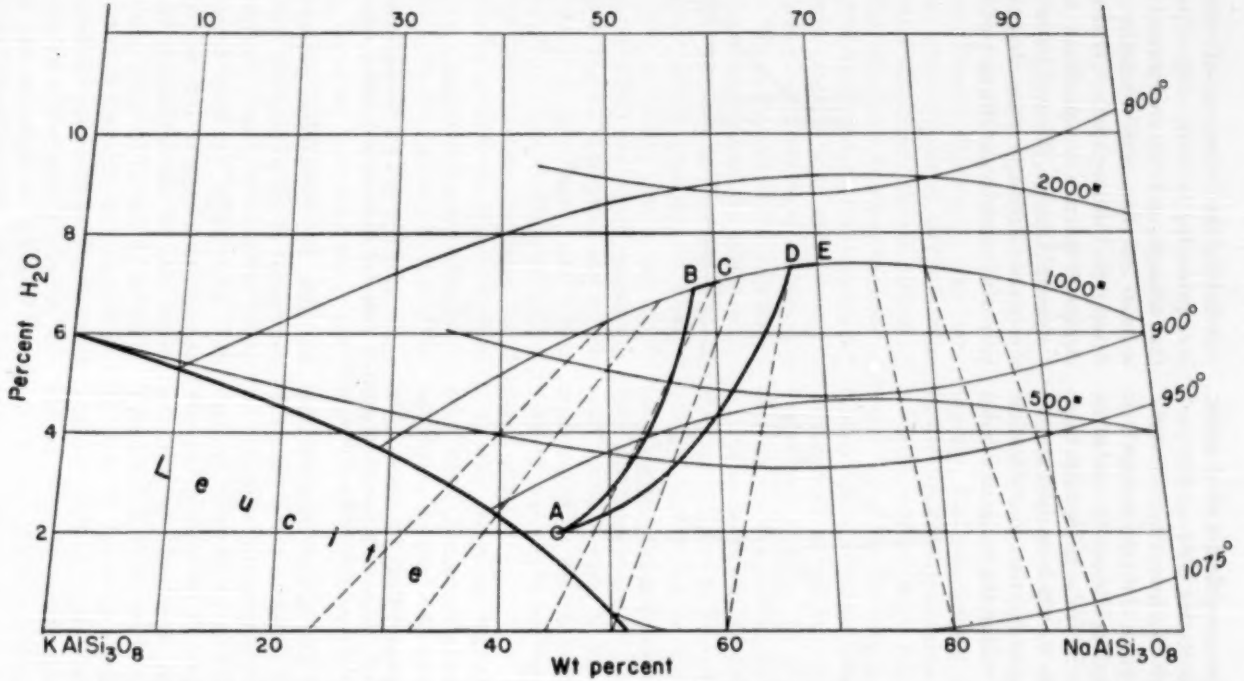

FIG. 7.—Portion of triangular diagram for the system $KAlSi_3O_8$-$NaAlSi_3O_8$-H_2O to illustrate courses of crystallization. Both the temperature saturation surface and the pressure saturation surface are projected on the triangle and are represented by isotherms and isobars, respectively. Pressures are indicated thus 500• = 500 kg/cm². Further details of method of construction of diagram in text.

desirable to know the feldspar proportions apart from water content, instead of drawing co-ordinates parallel to the other two sides of the triangle, lines radiating from the H_2O apex are drawn. With their aid the feldspar proportions represented by any point can be read directly, whatever the water content may be.

The isobars and isotherms are then plotted with the aid of the data on which the diagrams in figure 3 are based and also the determined water contents of the liquids (glasses), including Goranson's determinations. The significance of these curves is simply stated. The isobar of 1,000 kg/cm², for example, represents the composition of all liquids that are saturated with feldspar crystals and with water vapor at a pressure of 1,000 kg/cm², and an isotherm, for example, that for 950°, represents the composition of all liquids that are just saturated with feldspar crystals and water vapor at 950°. Tie-lines may now be added, joining the composition of any liquid represented by a point on an isobar with the particular feldspar in equilibrium with it. These tie-lines are given only for the 1,000 kg/cm² isobar. An attempt to show them for more than one isobar would result in a confusion of lines. Figure 3 supplies this information also, for horizontal lines on figure 3 are such tie-lines.

The diagram (fig. 7) is now ready for use to describe the course of crystallization of a liquid (magma) of any composition in the system. We shall bring out the principles involved by discussing crystallization of only one composition.

Composition *A* contains $NaAlSi_3O_8$ and $KAlSi_3O_8$ in the relative proportions 45:55 with 2 per cent H_2O. We shall assume that a liquid of that composition exists under an external pressure of 1,000 kg/cm² and at an elevated temperature, say 1,060°. Crystallization of this liquid will begin, if it is cooled, at approximately the temperature of the isotherm passing through *A*. It will not be exactly at the temperature of that isotherm, because, as will be recalled, the isotherms locate a saturation surface, the liquids being saturated with both water vapor and crystals. At 1,000 kg/cm² this liquid *A* is not saturated with water vapor, the saturation pressure being about 400 kg/cm². Accordingly, the temperature of initial crystallization at the higher pressure of 1,000 kg/cm² will be raised slightly above that indicated by the saturation surface. This effect of pressure in raising the crystallization temperature *in the condensed system* is small. Therefore, the liquid will begin to crystallize at a temperature slightly higher than that indicated by interpolation between isotherms on the saturation surface; but the actual difference is very small, and, for all practical purposes, isotherms on the saturation surfaces can be used.

Crystallization will therefore begin at approximately 1,000°. The first crystals formed will have the composition Ab_{14}; and, if the perfect equilibrium type of crystallization prevails, the first part of the course of crystallization is given by the curve *AB*. At *B*, which is a point on the 1,000 kg/cm² isobar, the liquid is now saturated with water vapor. The temperature, now indicated *precisely* by the isotherm through *B*, is, by interpolation, 850°. The position of the point *B* is determined by the tie-line (three-phase boundary) which passes through *A*. The mass now consists of liquid *B*, which contains nearly 7 per cent H_2O, and crystals of Ab_{40}. The proportion of liquid to crystals is approximately 30:70.

The assumed external pressure being 1,000 kg/cm², further abstraction of heat will result in boiling (vesiculation), ac-

companied by crystallization, and the composition of the liquid now moves along the isobar until the liquid has reached the point *C*, the temperature falling only a very little between *B* and *C*. At *C* crystallization will be complete, the mass having boiled dry and being made up entirely of crystals of composition Ab_{45}. Compared with the rate of cooling during earlier stages, that during the boiling stage is so small that it may be referred to as semi-isothermal. It is to be noted that, during the period of boiling, the water content of the residual liquid increases slightly, but the amount of residual liquid decreases markedly, being finally reduced to zero. The point indicating the total composition of the mass does not remain at *A* but migrates to the feldspar side of the diagram during the boiling.

With crystallization of the perfect fractionation type, initial formation of crystals will be as above, but thereafter the liquid will follow the course *ADE*, boiling beginning at *D*, a significantly lower temperature than *B*, and both boiling and crystallization being completed at *E*, which is the point of minimum temperature on the isobar (840°). The crystals in this case are continuously zoned, with compositions ranging from Ab_{14} to Ab_{70}. For a system similar to the present one the principles controlling crystallization by these two contrasted methods are discussed in a theoretical paper by Bowen (1941).

The assumed pressure of 1,000 kg/cm² prevailing during the crystallization of the liquid *A* corresponds with the weight of overburden at a depth of 2.2 miles in the earth, assuming a rock density of 2.7. With the aid of figure 7, crystallization for a pressure and, therefore, a depth twice as great (2,000 kg/cm²) can be described in a similar quantitative manner on the basis of our results. Indications are that at a pressure twice as great again, namely, 4,000 kg/cm², corresponding with a depth of 8.9 miles, the general situation will not be greatly different, that is, that the boiling-away of the water in the last residual liquid will take place in a manner similar to that described. The only difference will be that the last liquid will persist to a significantly lower temperature (estimated by extrapolation to be approximately 700°), and the content of water in the final liquid will probably be about 11 per cent. No reliance could be placed on more extended extrapolation, and it is futile to attempt to estimate the pressure (and depth), if any, at which critical phenomena would intervene and the final liquid (fluid), instead of boiling, would pass into the vapor (fluid) state without change of volume or discontinuity of any kind, including composition.

In the foregoing discussion of crystallization it has been assumed that the vapor escapes from the system entirely, in which case the crystalline feldspar cools down indefinitely without undergoing any change except the unmixing into two feldspars. If, however, the system were able to increase its volume—say by arching its roof—and thus retained the vapor as the filling of miarolitic cavities, then at a temperature around 400° or somewhat lower a liquid would form again. This is a liquid very rich in water (hydrothermal solution), but containing in solution some feldspar or, more probably, decomposition products of the feldspar, as the relations are probably incongruent. A notable recrystallizing effect may be expected of this liquid, but, of course, the retained vapor itself, existing in the temperature interval between that of magmatic liquids

and that of hydrothermal solutions, would also have a similar effect. The unmixing of the feldspar to give two feldspars is one phenomenon that would be notably facilitated by the presence of water in either form.

The reverse process, the heating of feldspar to a high temperature in the presence of some water, has some suggestive significance in connection with the problem of palingenesis. In a feldspar mass under a load of 1,000 kg/cm² and with sufficient pore space to contain 2 per cent water, the effects of heating would be the reverse of the cooling effects described if the volume relations are correct, that is, if the pore space is such that the 2 per cent water by weight fills or nearly fills it. The feldspar being assumed, of course, to have the same composition (Ab_{65}) as that whose crystallization was discussed, a magmatic liquid will first form at *C*, and further melting will be the exact reverse of crystallization. Even a minute trace of water *filling* the pore space in such a feldspar mass would induce the formation of a little magmatic liquid at *C*, the pressure again being assumed as 1,000 kg/cm². This liquid would, however, move along the isobar only a minute distance from *C* and then would leave the isobar and go to higher temperatures, with solution of more feldspar. In other words, no more than a very small quantity of liquid is formed unless the temperature is raised well above that of point *C*.

PETROLOGIC CONSIDERATIONS

The present investigation of crystallization equilibrium of the alkali feldspars confirms, with little room for doubt, the indications of dry-melt studies that these feldspars form a complete series of solid solutions of the type with a minimum-melting member. Many petrologists have considered this relation the most probable, solely from examination of the relations of the natural minerals. There is one major feature of the natural minerals that seems incapable of explanation on this basis. The potash-rich feldspars formed at high temperatures in rocks—the sanidines—give every indication of monoclinic symmetry even when they contain 40–50 per cent of the soda feldspar. High-temperature albite, on the other hand, gives as good indications of triclinic symmetry, and soda-rich alkali feldspars formed at high temperatures in rocks—anorthoclases—are usually so regarded also. We cannot resolve the difficulty but can only reiterate that the experimental results indicate complete solid solution. The crystals obtained synthetically are not of such quality as to enable one to decide on the crystal system.

Some consideration has already been given to the crystallization of the synthetic mixtures with water and need not be enlarged upon here. Suffice it to say that the existence of the lowest-melting composition at 65–70 per cent $NaAlSi_3O_8$ agrees with the indications of natural rocks; and, viewed in reverse, the relations of natural rocks approaching the synthetic mixtures in composition confirm the view that crystal-liquid equilibrium is the dominant control in the development of these rocks. It has long been argued that the presence of even moderate quantities of water in the natural magmas would completely change the relations found in dry melts, even to the extent of reversing the order of crystallization, and that studies of dry melts can therefore give no indication of the crystallization behavior of magma. No one having any real familiarity with equilibrium diagrams could adhere to

such views, and here we have a concrete illustration of the fact that the principal function of water is a fluxing one, in which capacity it is indeed very powerful, but it affects equilibrium positions progressively and only moderately when present in moderate amounts. The result is that, in investigated compositions containing water up to 10 per cent, crystallization equilibria are nearly the same as in the dry melts except that they go forward at much lower temperatures. Water does have an outstanding effect on one equilibrium, and that is on the incongruent melting of $KAlSi_3O_8$. Only about 6 per cent H_2O is sufficient to nullify this behavior, but its great fluxing power is probably at the root of this effect. Albite gives the same result eventually, but it requires 50 per cent albite to prevent formation of leucite. The difference is one of degree rather than of kind, but on this particular equilibrium the effect of water is indeed very great.

Those intergrowths of the alkali feldspars known as perthites, microperthites, and cryptoperthites have long been interpreted by petrologists as the result of the unmixing of a homogeneous solid solution. Our results confirm unmixing and supply information on its quantitative aspects, especially in indicating the minimum possible temperature of crystallization of a feldspar of a given composition as discussed on an earlier page.

It is not surprising that syenites and trachytes should be the principal abode of perthitic, and especially of microperthitic and cryptoperthitic, feldspars. In such magmas there is practically no constituent that induces a lowering of the melting temperatures of the feldspars except the water content, and this must be rather high, in order to lower the whole crystallization range to such a temperature that two feldspars will form throughout the period of primary crystallization. Therefore, we may expect that the bulk of the crystallization of such rocks took place at a temperature where a single homogeneous alkali feldspar is formed. This feldspar later unmixes to give a cryptoperthite, or a coarser intergrowth, according to the rate of cooling.

It should be noted, however, that if the pressure is high enough, that is, if crystallization takes place at sufficient depth in the crust, even a magma with a small initial percentage of water will eventually develop a residual liquid containing a notable amount. Thus a magma initially containing 1.2 per cent of water will have 12 per cent when the residual liquid is 10 per cent of the mass, unless notable amounts of water enter into hydrous minerals. Although our studies have not yet been carried to high enough pressures to permit a positive statement, this is probably about the maximum content of water possible in the low-melting feldspar mixture, and the temperature is probably down to a value where two distinct feldspars crystallize directly from the liquid.

At this stage the rate of heat flow from a deep-seated rock mass will be very small, and consequently the boiling-away of water and accompanying crystallization will proceed very slowly. The residual liquid, amounting at first to 10 per cent and slowly decreasing from that value, will be present for an excessively long period. By virtue of its high water content, it is a very active liquid and may be expected to effect complete recrystallization of the early-formed feldspar, with formation of those with which it is in equilibrium. To be sure, if there is present only the feldspars and water, the temperature, while liquid persists, will never go far down into the range of

two feldspars, that is, the feldspars will not be of extreme composition, but in the natural system residual liquids will have potent fluxes in addition to water, and this recrystallization may be expected to be carried to significantly lower temperatures with the aid of residual liquid. Through this action the typical deep-seated syenite may be formed. It should be realized, too, that at very high pressures approaching critical end-points the vapor phase formed is not greatly different in composition from the liquid phase, as figures 5 and 6 show, and this vapor phase, filling minute interstices, will also induce recrystallization even after the exhaustion of all liquid.

It is such vapors and the vapors boiling off at earlier stages that introduce material into surrounding rocks, with resultant "granitization" of these rocks.

The fact that, in compositions containing feldspars and water, liquids do not get far down into the region of two feldspars is a restriction that does not apply to compositions with an excess or a deficiency of silica. The granitic composition resulting, on the one hand, and the nepheline syenite composition, on the other, require lesser concentrations of water to lower their crystallization temperatures into the region of two feldspars, but even these may be expected to have early feldspars that are no longer stable at the temperatures approaching final crystallization and are consequently recrystallized by the late residuals in the manner described.

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THE ALKALI-FELDSPAR JOIN IN THE SYSTEM $NaAlSiO_4$-$KAlSiO_4$-SiO_2[1]

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ABSTRACT

A study of the alkali-feldspar join $KAlSi_3O_8$-$NaAlSi_3O_8$ in the system $NaAlSiO_4$-$KAlSiO_4$-SiO_2 shows an unbroken series of solid solutions between these alkali feldspars with a minimum on the melting and freezing curves at 1,063° ± 3° C. at 35 weight per cent potash feldspar. All alkali-feldspar compositions with more than 49 weight per cent potash feldspar melt incongruently and have leucite crystals present, which disappear only at higher temperatures. A revised equilibrium diagram for the system $NaAlSiO_4$-$KAlSiO_4$-SiO_2 is given; the data on which it is based will be published in full later.

INTRODUCTION

Because of the widespread occurrence of feldspars in rocks, the stability relations of the alkali feldspars are of extreme importance and interest. During the years between 1929 and 1941 the phase-equilibrium relations, at temperatures at which a liquid phase was present, were determined in the ternary systems Na_2O-Al_2O_3-SiO_2 and K_2O-Al_2O_3-SiO_2 by J. F. Schairer and N. L. Bowen (1947*a*). Concurrently with the studies of these two systems, the melting relations in $NaAlSiO_4$-$KAlSiO_4$-SiO_2 were investigated. This last investigation is still not completed because of (1) difficulties with crystallization in the dry, viscous, siliceous melts, (2) the slow rate of attainment of equilibrium between crystals and liquids, (3) difficulties with the precise determination of the composition of some of the solid crystalline phases, and (4) interruption of these studies by World War II.

Soda feldspar ($Na_2O \cdot Al_2O_3 \cdot 6SiO_2$ or $NaAlSi_3O_8$) is a ternary compound in the system Na_2O-Al_2O_3-SiO_2. Greig and Barth (1938) showed that this compound melts congruently at 1,118° ± 3°. This was confirmed by Schairer and Bowen (1947*a*), who also showed the melting relations of soda feldspar in the presence of an excess of each of its components.

Similarly, potash feldspar ($K_2O \cdot Al_2O_3 \cdot 6SiO_2$ or $KAlSi_3O_8$) is a ternary compound in the system K_2O-Al_2O_3-SiO_2. Morey and Bowen (1922) showed that three samples of natural potash feldspars with compositions low in soda feldspar and a pure synthetic potash feldspar (prepared hydrothermally) melted incongruently to a liquid more siliceous than feldspar plus crystals of leucite ($K_2O \cdot Al_2O_3 \cdot 4SiO_2$ or $KAlSi_2O_6$). These leucite crystals disappeared only when the temperature was raised to about 1,530°. They placed the incongruent melting temperature of pure potash feldspar at about 1,170°. Schairer and Bowen (1947*a*) confirmed these results and placed the incongruent melting temperature of potash feldspar at 1,150° ± 20° and also showed the melting relations in the presence of an excess of each of its components. They found that dry melts of compositions between potash feldspar and silica are extremely viscous and that most of them, except those with cristobalite liquidus temperatures above about 1,350° (those below the cristobalite ⇌ tridymite inversion at 1,470° ± 10° are

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metastable) could not be crystallized dry. However, they were able to present the relations between potash feldspar and silica in the binary system leucite-silica (Schairer and Bowen, 1947*b*) by a short extrapolation from data on the ternary compositions, K_2O-Al_2O_3-SiO_2, with small excess potash over potash feldspar–silica compositions. These ternary compositions were much less viscous than binary leucite-silica compositions and were amenable to crystallization and study by the method of quenching.

THE SYSTEM $NaAlSiO_4$-$KAlSiO_4$-SiO_2

Schairer and Bowen (1935) published a preliminary report on the equilibrium relations of feldspathoids, alkali feldspars, and silica in the system $NaAlSiO_4$-$KAlSiO_4$-SiO_2 and gave two preliminary diagrams (1935, figs. 1 and 2) for this system, the one showing the fields of the several primary crystalline phases and the other the compositions studied up to that time, with some of the isotherms. No field for orthorhombic $KAlSiO_4$ was shown because data for that area were lacking at that time. Very little data were available then for compositions richer in silica than the alkali-feldspar join. Some of the temperatures indicated on these preliminary diagrams were too high because sufficient time had not been allowed for the attainment of equilibrium between crystals and liquid in these viscous melts.

The revised equilibrium diagram for the system $NaAlSiO_4$-$KAlSiO_4$-SiO_2 is given here as figure 1. The data on which it is based will be published in full in a later paper. Solid black circles indicate the compositions that have been prepared and studied. Those compositions shown as solid black triangles have been prepared, but no quenching data for them have been obtained because they have not yet been crystallized, even though these dry glasses have been held at appropriate temperatures for as long as 5 years. Heavy solid curves (or a dashed curve if the data are not complete) are the boundary curves between the fields of the several primary crystalline phases. Arrows on these curves indicate the direction of falling temperature. Isotherms are shown as light lines, with the temperature of the isotherm indicated. The shape of the isotherms in the alkali-feldspar field indicates the probability of a temperature minimum on the boundary curve between the fields of alkali feldspar and tridymite. No minimum is shown here, because quenching data are entirely lacking for this region for reasons already given.

The alkali-feldspar join divides the ternary system into two portions. Those compositions between it and the silica apex approach compositions of simplified dry granitic rock magmas, where the ultimate products of crystallization are a feldspar and the appropriate form of silica. Those compositions between the alkali-feldspar join and $NaAlSiO_4$-$KAlSiO_4$, the base of the triangle, approach compositions of possible simplified alkaline rock magmas, where the ultimate products of consolidation are nepheline (not pure $NaAlSiO_4$ but hexagonal mix crystals of $NaAlSiO_4$, $KAlSiO_4$, and alkali feldspar), alkali feldspar, and leucite, except for extreme compositions near the $NaAlSiO_4$-$KAlSiO_4$ base which crystallize to a nepheline alone or those near the $NaAlSiO_4$–soda feldspar side, which may crystallize to a nepheline and a feldspar with no leucite in the completely crystalline product.

THE ALKALI-FELDSPAR JOIN

Schairer and Bowen (1935) published a preliminary diagram for the join $KAlSi_3O_8$-$NaAlSi_3O_8$ and interpreted the data (admittedly incomplete) as showing an unbroken series of solid solutions between these two alkali feldspars, with a minimum on the melting and freezing curves and the further complication of incongruent melting for all the feldspar compositions with more than about 49 per cent of potash feldspar. Since that time additional alkali-feldspar compositions have been prepared, so that compositions at each 5 per cent interval of alkali-feldspar proportions were available for study.

The revised equilibrium diagram for the alkali-feldspar join is given here as figure 2. It differs in no essentials from the preliminary diagram of Schairer and Bowen (1935, fig. 3). Heavy lines (or heavy dashed lines if the data are less precise) refer to binary equilibrium. All curves that refer to ternary equilibrium are shown as lighter lines (or lighter dashed lines). These lighter lines give only the temperature at which phase changes occur in a preparation of a given total composition. They do not give the composition of all phases concerned in the equilibrium, since these relations can be shown only with the aid of a ternary diagram. Determined points are shown

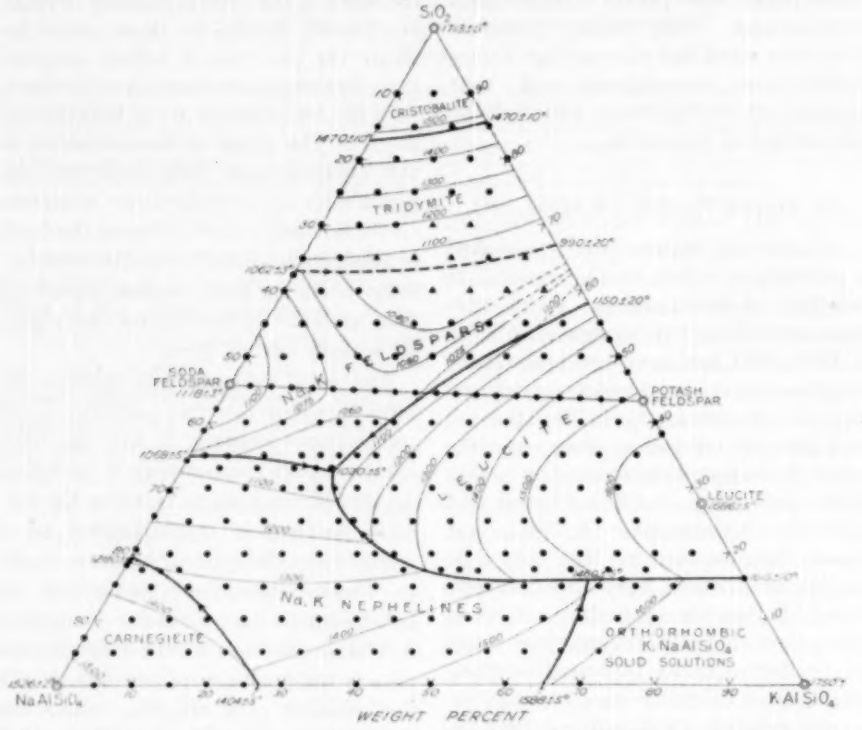

FIG. 1.—Revised equilibrium diagram for the system $NaAlSiO_4$-$KAlSiO_4$-SiO_2, showing position of the alkali-feldspar join. Data on which the ternary diagram are based will be published in full in a later paper.

as open circles. The quenching data on which they are based are given in table 1. *AD* (fig. 2) is the liquidus curve for leucite. *DEF* (fig. 2) is the liquidus curve for alkali feldspars and shows a temperature minimum at *E* (fig. 2) at 1,063° ± 3°, with the composition 35 per cent $KAlSi_3O_8$ and 65 per cent $NaAlSi_3O_8$. The temperature of *CD* (fig. 2) is 1,078° ± 3°. No data are available on the dry melts to fix the composition of *C* (fig. 2), and for this reason the relations near this point are shown as dashed curves.

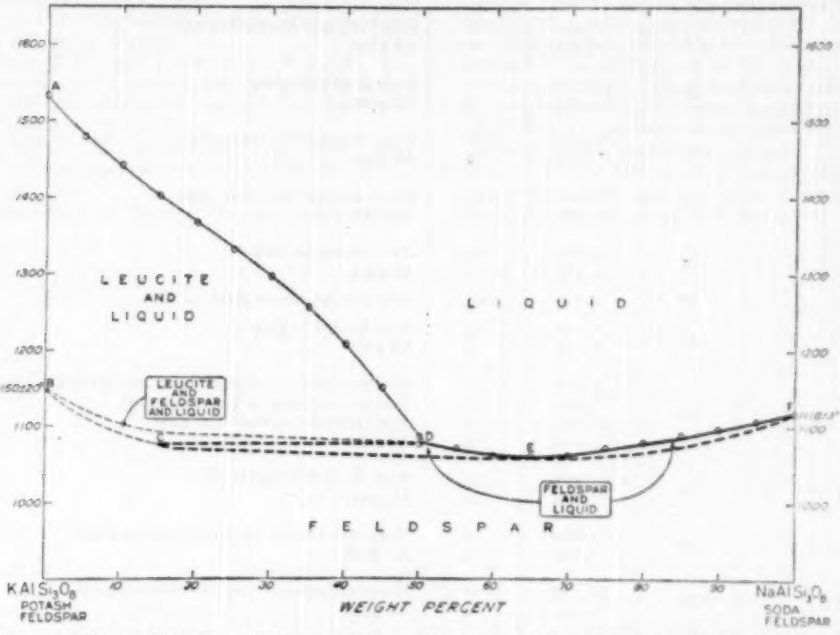

FIG. 2.—The alkali-feldspar join. Heavy solid or dashed curves refer to binary equilibrium. Light solid or dashed curves refer to ternary equilibrium. Open circles indicate determined points.

All alkali-feldspar compositions and particularly those with more than 50 per cent of potash feldspar give viscous melts that are difficult to crystallize dry, and in these crystalline or partly crystalline melts equilibrium between crystals and liquid is attained very slowly. It was possible to determine liquidus temperatures within narrow temperature limits by observing temperatures at which a small amount of tiny crystals grew in the melt and above which they were dissolving. On the other hand, it was not possible to determine the beginning of melting, since none of these alkali-feldspar compositions could be completely crystallized dry and since, even if crystallized hydrothermally, no accurate value for beginning of melting can be obtained because the crystals must be heated above their temperature of beginning of melting for a week or several weeks before observable melting (glass under the microscope) can be recognized.

Thus we see that a study of dry melts can yield only a limited amount of information about the alkali feldspars. Such a study does show the general relations at temperatures at which a liquid

TABLE 1

RESULTS OF QUENCHING EXPERIMENTS

COMPOSITION (WEIGHT PER CENT)		TEMP. (° C.)	TIME (DAYS)	RESULTS OF MICROSCOPIC EXAMINATION
$KAlSi_2O_6$	$NaAlSi_2O_6$			
95	5	1,476	1	Rare leucite in glass
		1,480	1	All glass
90	10	1,440	2	Small amount leucite in glass
		1,445	2	All glass
85	15	1,400	6	Small amount leucite in glass
		1,405	6	All glass
80	20	1,365	7	Rare leucite in glass
		1,370	7	All glass
75	25	1,330	7	Small amount leucite in glass
		1,335	7	All glass
70	30	1,295	14	Small amount leucite in glass
		1,300	14	All glass
65	35	1,255	14	Rare leucite in glass
		1,260	10	All glass
60	40	1,210	14	Very rare leucite in glass
55	45	1,150	14	Rare leucite in glass
		1,155	14	All glass
50	50	1,077	30	Moderate amount of leucite and rare feldspar in glass
		1,080	35	Moderate amount of leucite in glass
		1,085	35	Small amount of leucite in glass
		1,090	14	All glass
45	55	1,070	27	Rare sharp feldspar in glass
		1,075	14	All glass
40	60	1,060	21	Moderate amount sharp feldspar in glass
		1,065	14	All glass
35	65	1,060	21	Moderate amount sharp feldspar in glass
		1,065	14	All glass
30	70	1,060	21	Small amount sharp feldspar in glass
		1,065	14	All glass
25	75	1,070	21	Rare sharp feldspar in glass
		1,075	14	All glass
20	80	1,075	21	Moderate amount sharp feldspar in glass
		1,080	21	Very rare sharp feldspar in glass
		1,085	14	All glass
15	85	1,085	14	Moderate amount sharp feldspar in glass
		1,090	14	All glass
10	90	1,095	14	Rare sharp feldspar in glass
		1,100	14	All glass
5	95	1,105	14	Small amount sharp feldspar in glass
		1,110	14	All glass

phase is present. It does not tell us about the possibility of unmixing at lower temperatures or the possibility of inversions to other crystalline forms. My colleagues, N. L. Bowen and O. F. Tuttle, are engaged in a study of feldspar systems with water in which the melts are much less viscous and equilibrium is attained much more rapidly, even though the temperatures are considerably lower. Their results, some of which are presented in companion papers herewith, give us a more precise picture of the mineralogical and geological relations of feldspars.

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GALLIUM AND GERMANIUM SUBSTITUTIONS IN SYNTHETIC FELDSPARS[1]

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ABSTRACT

Compounds of gallium and germanium isostructural with the feldspars have been prepared, and some of their crystal-chemical properties are here described. In the Na and K feldspars, Al^{+++} and Si^{++++} can be completely replaced by Ga^{+++} and Ge^{++++}; but, in pure Ca feldspar, complete substitutions cannot be made. The use of these substitutions as a means of obtaining information on the feldspars is discussed.

One of the principal aims of this line of investigation is to obtain information on substitutional disorder in alumino-silicates. It is difficult to locate specific positions of Al and Si atoms in many silicates because of the approximately equivalent X-ray scattering power of these two atoms. The use of a heavier-element replacement should produce more definite information on the degree of substitutional disorder, and it is hoped that these Ga and Ge feldspars will help in the interpretation of some of the problems of the natural minerals.

INTRODUCTION

Rather widespread interest in the problem of order-disorder in minerals has developed in the past few years. Very little has been done to date on this problem as it pertains to minerals, but a sizable literature on the general subject of order-disorder relations in crystals has accrued since 1916, when Tammann's original observations in the Cu-Au system appeared. One of the aims of the present investigation is to obtain information on one type of disorder that may occur in alumino-silicates, namely, substitutional disorder between silicon and four-co-ordinated aluminum atoms. A description of the synthesis of some model feldspar structures and some discussion of their crystal-chemical properties—the first phase of this investigation—is here presented. The specific relationship of the synthetic compounds to the broader problem of feldspar disorder will be the subject of a later paper.

Inasmuch as this paper does not deal with the general problem of disorder in crystals, earlier work will not be summarized in detail. Only the more important contributions, particularly those that apply to the feldspars, will be mentioned. In an excellent treatment of the problem of disorder in alumino-silicates, Schlecht (1946) reviews the history of the order-disorder concept and gives abundant references. Mineralogical disorder was also discussed by Buerger (1948), who points out in particular the relation between disorder and exsolution in solid solutions.

Jaeger, Westenbrink, and Van Melle (1927), in determining the structure of ultramarine, were the first to show that aluminum and silicon atoms could replace each other in a tetrahedral framework. F. Machatschki (1928) correctly proposed an interpretation of the feldspar structure based on linked Si-O and Al-O tetrahedra, with the cations interstitially located in the framework. Barth and Posnjak (1931) clearly expressed the concept of random arrangement of particular atoms in a structure, based on work done with spinels. They described structures of this type as having "variate atom equipoints." In 1933 W. H. Taylor,

[1] Manuscript received April 21, 1950. Presented at the November, 1949, Annual Meeting of the Geological Society of America.

working with sanidine, was the first to solve satisfactorily a feldspar structure. He could not distinguish Al from Si atoms, however, but recognized that "... the tetrahedra of oxygen atoms around Al atoms will probably be larger than those around silicon atoms...." He attributed some of his discrepancies in observed versus calculated intensities as possibly due to not taking this into account. He found four $KAlSi_3O_8$ "molecules" per unit cell and discovered that the sixteen (Al and Si) atoms were divided into two structural groups of eight. Not considering the possibility of disorder, he stated that the two groups, eight "Si_1" and eight "Si_2," must really be split into four groups, of which three contain four Si each and one contains four Al. This division was based on the assumption that sanidine did not show monoclinic holohedral symmetry. Barth, in 1934, applied his concept of variate atom equipoints to the potash feldspars. He questioned Taylor's symmetry classification of orthoclase, claiming that all previous crystallographic measurements show it to be holohedral. He thought that in microcline the four Al atoms concentrate in two twofold positions, resulting in a triclinic structure. The monoclinic feldspars, adularia and sanidine, were pictured as having Si and Al as variate atoms. His theory as to the relationship between adularia and sanidine was admitted to be more uncertain, but he stated that some "interchange of silicon and aluminum atoms takes place." Barth thus proposed an explanation for feldspar polymorphism based on order-disorder relations.

Taylor, Darbyshire, and Strunz (1934) published a more general paper on the feldspars, that came out at the same time as Barth's. Here sanidine is assumed to be holohedral, which implies that Al atoms are randomly distributed (at least within one of the two Si groups). The structures of albite and anorthite are also presented, with the observation that the c-axis of anorthite is twice that of albite; this effect was ascribed to the different environments of Ca and Na atoms rather than to Al-Si differences. In 1940 Chao and Taylor presented a more detailed study of the plagioclase feldspars, which, in addition to the above-mentioned effect, showed more complex structures of the superlattice type in the intermediate plagioclases. The author's interpretation of their X-ray data is that the intermediate plagioclase is formed of alternating stacked layers (along the c-axis) of anorthite or anorthitic feldspar and albite or albitic material, each structural unit being a few unit cells thick. They thus do not consider that the plagioclases form a true, structurally complete, isomorphous series.[2] It should be remembered, however, that, because of the change in Al:Si ratio from albite to anorthite (1:3 to 1:1), a greater opportunity for more complex disorder exists than in the feldspars with a fixed Al:Si ratio.

A detailed quantitative X-ray study of orthoclase was made by Chao, Hargreaves, and Taylor in 1940. The distances from the Si and Al atoms to the O atoms are given; Si_1 and Si_2 types again are differentiated. The distances from the Si_1 groups of eight atoms to the different oxygen positions are 1.66, 1.67, 1.69, and 1.70 Å. The Si_2 group shows but two different Si-O spacings for the eight atoms—1.57 and 1.60 Å. The authors call attention to the difference between the two types of tetrahedra. Inasmuch as the AlO_4 group is larger than

[2] Chao and Taylor do not discuss the possibility of a complete isomorphous series at high temperatures and an incomplete series at room temperature. This point is treated by Buerger (1948).

the SiO_4 group, they conclude that the Si_1 type, showing smaller spacings, contains only Si atoms, whereas the Si_2 group contains four Si and four Al atoms. This conclusion is the same as that reached by Taylor in 1933, on the basis of qualitative X-ray work. It is further stated that the Al distribution in the Si_2 group is random, as there is no evidence of any reduction of the symmetry from holohedry. The authors also state: "It may be that the distribution of the aluminum ions among the tetrahedral groups is of primary importance in the process of 'sanidinization' revealed by the irreversible change in optic axial angle."

In a very recent (1949) paper by Cole, Sörum, and Kennard the symmetry relations with respect to Al-Si distribution in orthoclase and sanidine are more fully discussed. The atomic parameters of sanidine were determined with great precision. They find such a small variation in bond lengths around the two tetrahedral groups (Si_1 and Si_2) that the Al atoms must be completely disordered in this material. These authors also re-examined the 1940 work of Chao, Hargreaves, and Taylor and show that the accuracy is not adequate to justify their conclusion regarding Al atom distribution. To establish whether or not significant differences exist in the size of the tetrahedral groups, it is pointed out that orthoclase must be re-examined with the accuracy of the present investigation. The status of the disorder problem in the feldspars, in so far as present published X-ray work is concerned, thus appears to show complete Al-Si disorder in sanidine; nothing definite has been proved as to the situation in the other potash feldspars or in the plagioclases. Any completely ordered arrangement of Al atoms in a *monoclinic* feldspar would, however, produce a reduction in symmetry from that assumed.[3]

K. J. Murata (1946) discusses the effect of acids on silicates with relation to their structure. Their behavior is ascribed to the size of the Si-O units that make up the structure or to the fact that the structure can be broken down by virtue of weak points produced, for example, by Al atoms. His hypothesis is that small Si-O units dissolve in HCl or HNO_3 to yield gelatinous silica, whereas large units produce insoluble separated silica. W. G. Schlecht (1946) treats the general problem of random versus ordered arrangement of a two-component array and considers the size of clusters that would result, depending on the degree of randomness of the array. The problem of the alumino-silicates is discussed, and Schlecht states: "In mixtures with compositions like those of aluminosilicate minerals, the number of small clusters becomes negligible as complete randomness of arrangement is approached, and it is concluded on the small-cluster hypothesis of gelatinization of silicate minerals that the arrangement of aluminum and silicon atoms must be very symmetrical with respect to each other, otherwise clusters too large to gelatinize would be formed when the minerals are decomposed." Murata points out that, with two exceptions, the silicates with three-dimensional structures gelatinize if the Al:Si ratio is 2:3 or greater; the more siliceous structures separate insoluble silica (or are not broken down at all). The earlier work (1944) of Wichers, Schlecht, and Gordon on the decomposition of plagioclase feldspars in sealed tubes in acid at 300° C. is also of interest in the light of the foregoing discussion.

[3] See discussion by Fritz Laves, this issue of the *Journal*.

SIGNIFICANCE OF Ga AND Ge REPLACEMENTS IN SYNTHETIC FELDSPARS

The scattering factor of X-rays by atoms is a function of atomic number. The atomic number of Al is 13; of Si, 14. Thus, because of similar X-ray scattering powers, the relative position of Al atoms and Si atoms in alumino-silicate networks cannot generally be distinguished. If the Si-Al order-disorder relations in this most important group of minerals could be evaluated, many mineralogical and petrological problems might be clarified.

If, in an alumino-silicate, the Al atoms could be replaced by an element of significantly different atomic number or if Si could be replaced in like fashion, it would be possible, by means of X-ray diffraction, to detect positional differences of the two elements. The elements replacing Si or Al would, of course, have to be so similar in other respects that the original structure would be maintained. In addition to the possibility of detecting order-disorder relations, there is the opportunity to observe other interesting crystal-chemical effects produced by controlled substitutions of this sort. The pairs aluminum-gallium and silicon-germanium are crystal-chemically perhaps the most closely related pairs of elements. In ionic crystals—and silicates are predominantly ionic—radius and charge are the most important properties governing crystal structures. These properties of Ga and Ge are compared with those of Al and Si in table 1.

If an alumino-silicate structure can be duplicated by replacement of the Al or Si with other elements, Ga and Ge are the most likely elements; the charges are the same, and the radii are very similar.[4] The large differences in atomic number between Al and Ga and between Si and Ge are also very favorable for detection by means of X-ray diffraction.

Observations of any disorder phenomena in such "model" structures might be of value in the interpretation of their natural counterparts. In addition to the use of X-ray diffraction, other techniques might also be used: for example, the chemical approaches as applied to silicates by Murata (1946) and by Wichers, Schlecht, and Gordon (1944). The change in relative bond strengths (i.e., Al-O vs. Si-O) produced by such substitutions might be important in this respect.

TABLE 1

Element	Ionic Charge	Ionic Radius (Å)*	Atomic Number
Al	3+	0.57	13
Ga	3+	0.62	31
Si	4+	0.39	14
Ge	4+	0.44	32

* Radii given for sixfold co-ordination.

Synthetic feldspars have been made with 100 per cent substitutions of Ga for Al, Ge for Si, and, to complete the series, both Ga and Ge substituted for Al and Si. A stable lime feldspar structure does not appear to form with complete Ga, Ge, or Ga and Ge substitutions, but the alkali feldspar structures do. In the following sections these new compounds with feldspar structures are discussed.

EXPERIMENTAL PROCEDURE

Mixtures of the desired compositions were prepared by fusing the carefully mixed pure oxides or carbonates to a glass, in platinum crucibles. The glasses

[4] For a more complete discussion of these similarities and for a description of some Ge compounds isostructural with Si compounds see the 1931 papers of V. M. Goldschmidt.

were crushed, mixed, and re-fused until optically homogeneous. Three fusions generally produced a homogeneous melt, except in the case of the Ga equivalent of orthoclase, $KGaSi_3O_8$. Here, because of very high viscosity, six fusions were necessary to completely melt and homogenize the mixture. Loss of K_2O by volatilization must also be corrected for in this composition. Volatile loss of GeO_2 in the mixtures that require high fusion temperatures is also a problem, as is attack and contamination of the platinum crucibles when melting the potash compounds.

The Ga and Ge used in this investigation were very pure,[5] and were both obtained from the research department of the Eagle-Picher Lead Company. GeO_2 was supplied and used as such; Ga_2O_3 was prepared from metallic Ga by dissolving the metal in nitric acid and evaporating to dryness, the nitrate being decomposed to the oxide by heating. The other batch ingredients—Na_2CO_3, $KHCO_3$, $CaCO_3$, Al_2O_3, and SiO_2—were all of very high purity.

The crystalline materials were prepared either by devitrification of the glasses at some convenient temperature below the melting point of the crystalline phase dealt with or by hydrothermal crystallization, the latter done either in Morey-type bombs or in a device of the type described by Tuttle (1948). The possibility of structural variation as a function of temperature of crystallization is not considered in the present paper. In several cases, to be described later, mixtures of the batch ingredients were reacted directly in the solid state rather than being first melted to a glass. This technique may be of more value in later phases of this investigation concerned with the afore-mentioned structural variation. It is at times expedient to avoid the possibility of any inherited structure from the high-temperature or glassy state.

All melting points were determined by the well-known quenching technique. The quenched materials were all examined under the microscope.

[5] Spectrographic analyses, made by Eagle-Picher, are as follows:

Impurity	Estimated Average Analysis (Per Cent)
GeO_2	
Silicon	0.00014
Magnesium	0.00044
Copper	0.00013
Calcium	0.0014
Lead	0.0005
Gallium Metal	
Silicon	0.0001
Magnesium	0.0004
Copper	0.007
Calcium	0.01
Lead	0.005
Silver	0.00003

THE ALKALI Ga AND Ge COMPOUNDS

SODA FELDSPAR STRUCTURES

The Ga and Ge counterparts of albite, $NaAlSi_3O_8$, are:

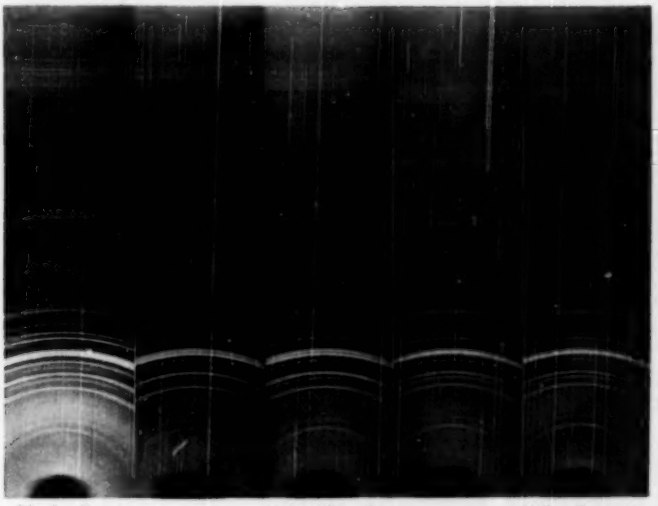
$NaGaSi_3O_8$ —"gallium albite"[6]
$NaAlGe_3O_8$ —"germanium albite"
$NaGaGe_3O_8$—"gallium-germanium albite"

The fact that these compounds have the albite structure is shown by the X-ray powder photographs. Plate 1, *A*, com-

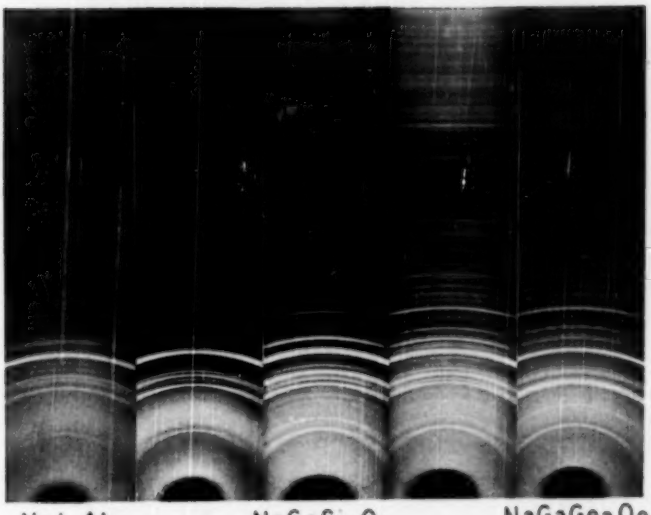
[6] This nomenclature, or the shortened forms, Ga-albite, Ge-albite, and Ga-Ge-albite, will be, in general, used to designate these compounds. The same prefix-type designation will be used in conjunction with "orthoclase" and "anorthite" for the K and Ca compounds.

PLATE 1

A, X-ray powder photographs of the soda compounds. FeKa radiation.
B, X-ray powder photographs of the potash compounds. FeKa radiation.

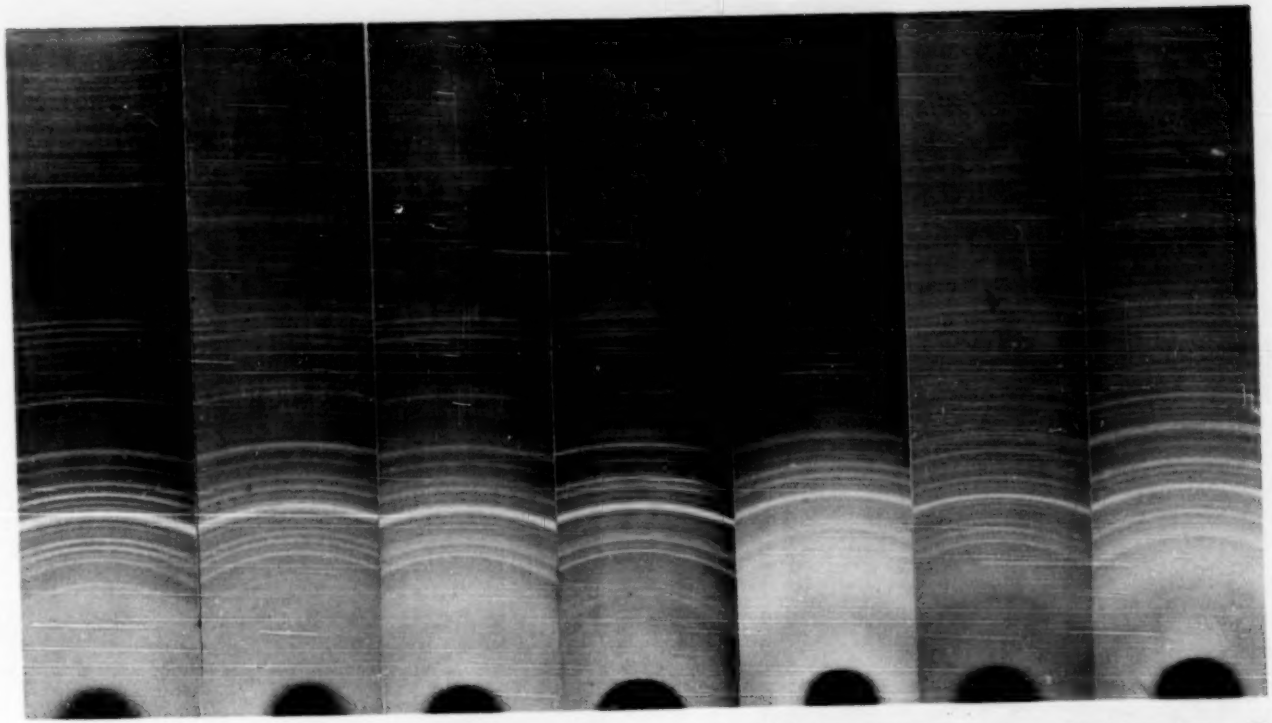


A



B

X-ray powder photographs of the alkali compounds



X-ray powder photographs of the lime compounds Fe K α radiation

pares the powder diffraction patterns of these compounds with pure natural and synthetic albites. These photographs are here introduced merely to show the general structural relations. As mentioned above, no interpretation of X-ray diffraction differences that may be significant to the disorder problem are discussed in this paper.

The optical properties, as far as they have been investigated, are those that would be expected to be produced by substitution of the heavier elements in the albite structure. The refractive indices are considerably higher than those of normal albite; the birefringence is rather low. Twinning is common. Table 2 lists some of the optical data determined on the albite series, with normal albite included for comparison. Variation of refractive indices in some of the compositions has been observed and may be a function of the temperature of crystallization or of dry versus hydrothermal formation. Additional optical data (sign, $2V$, etc.) are not presented because of the yet uncertain relations.

TABLE 2

REFRACTIVE INDICES OF THE SODA FELDSPARS AND THEIR GLASSES

Refractive Index	$NaAlSi_3O_8$	$NaGaSi_3O_8$	$NaAlGe_3O_8$	$NaGaGe_3O_8$
Glass	1.489	1.519	1.592	1.636
Crystals				
α	1.525	1.552	1.606	1.638
γ	1.536	1.558	1.619	1.654

The difference between the refractive indices of the crystals and their corresponding glasses becomes smaller in the order Ga-albite, Ge-albite, and Ga-Ge-albite; the difference becomes quite small in $NaGaGe_3O_8$. This effect will be discussed in the case of the lime compounds, where it is quite pronounced.

Melting points of the soda compounds are as follows, again compared with normal synthetic albite:

$NaAlSi_3O_8$ —1,118° C.
$NaGaSi_3O_8$ —1,015°
$NaAlGe_3O_8$ —1,067°
$NaGaGe_3O_8$— 952°

All melt congruently, as does normal albite. Table 3 lists the pertinent thermal data for the melting-point determinations. A melting range of approximately 10° is observed in Ge-albite and a larger range with the Ga-albite used. The interval in Ge-albite is probably due to the material being somewhat off-composition; an uncorrected volatile loss, probably of Na_2O, GeO_2, or both, was observed when the original glass was melted. The Ga-albite glass, being very viscous, was crystallized hydrothermally. The resulting crystals, used for melting-point determinations, contained some unknown impurity phase or phases (possibly due to differential solubility of the material). This, coupled with the inherent difficulty of determining melting points of very viscous materials, would account for the observed melting interval. The top of the interval was taken to be the melting point in both cases, and the error is probably not large.

Glass of the albite composition is very viscous and, as mentioned above, Ga-albite glass is also. Crystallization from a dry melt is exceedingly slow. Hydrothermal crystallization proceeds quite rapidly, however, even at temperatures as low as 400°. The very high viscosity and consequent difficulty of dry crystallization is not found in the Ge-containing albites. Ge-albite and Ga-Ge-albite crystallize readily from the dry melt. The high degree of polymerization of SiO_4 groups present in silica-rich melts (as evidenced from their high viscosity) is

not so apparent in the Ge-containing counterparts of these melts. Greater strength of the Si-O bonds as compared to the Ge-O bonds may not be the only factor involved in this difference; the possibility exists that Ge has a statistical co-ordination greater than 4 in the melt, which would reduce the degree of polymerization.

The melting point of Ga-albite is appreciably lower than that of Ge-albite. The melting point of Ge-albite is approximately 50° C. below that of normal albite, whereas that of Ga-albite is over 100° C.

TABLE 3

THERMAL DATA FOR THE SODA FELDSPARS

Composition	Original Condition	Time	Temp. (° C.)	Final Condition
$NaGaSi_3O_8$	Crystals (hydrothermally crystallized, rather impure)	4 days	900	Xln Ga-albite+interstitial glass
		3 days	1,001	Tiny Xls of Ga-albite in glass
		24 hr.	1,010	Very sparse tiny Xls in glass
		24 hr.	1,019	All glass
$NaAlGe_3O_8$	Crystals	3½ hr.	1,061	Xln Ge-albite+sparse interstitial glass
		5 hr.	1,065	Glass, sparse Xls Ge-albite
		24 hr.	1,069	All glass
$NaGaGe_3O_8$	Glass	2 hr.	950	Xls Ga-Ge-albite (rare glass?)
		16 hr.	955	All glass

THE POTASH FELDSPARS

The potash compounds equivalent to orthoclase, $KAlSi_3O_8$, are:

$KGaSi_3O_8$ —"gallium orthoclase"
$KAlGe_3O_8$ —"germanium orthoclase"
$KGaGe_3O_8$—"gallium-germanium orthoclase"

Two distinct modifications of $KAlGe_3O_8$ and of $KGaGe_3O_8$ have been observed. The two modifications so far found are readily distinguished by means of their X-ray patterns, but the refractive indices are the same or very similar. Only one modification of $KGaSi_3O_8$ has been found to date, corresponding in X-ray pattern to the high-temperature forms of $KAlGe_3O_8$ and $KGaGe_3O_8$ and to normal orthoclase.

The general term "orthoclase" is used with no implication as to whether adularia, sanidine, or, for that matter, microcline structural types are involved. The X-ray powder diffraction pictures of these potash compounds are shown in plate 1, *B;* a pure natural orthoclase and synthetic orthoclase are included for comparison. Only the high-temperature forms of $KAlGe_3O_8$ and $KGaGe_3O_8$ are shown. The isostructural relation of these compounds to normal potash feldspar is evident. The similarity of all the X-ray photographs in the case of the potash compounds is indeed striking.

TABLE 4

REFRACTIVE INDICES OF POTASH FELDSPARS AND THEIR GLASSES

Refractive Index	$KAlSi_3O_8$	$KGaSi_3O_8$	$KAlGe_3O_8$	$KGaGe_3O_8$
Glass	1.488	1.513	1.578	1.617
Crystals				
α	1.518	1.533	1.595	1.615
γ	1.526	1.539	1.590	1.628

The refractive indices of the "orthoclase" group are presented in table 4. It should be pointed out that, as in the case

of the Na feldspars, refinement of these data may be necessary, and, in addition, a more complete study will be made. The need for refinement is not based on a desire to obtain greater precision of fixed values but to observe variations as a function of conditions of crystallization.

The difference between the refractive indices of the crystals and their glasses

TABLE 5

THERMAL DATA FOR THE POTASH FELDSPARS

Composition	Original Condition	Time	Temp. (° C.)	Final Condition
$KGaSi_2O_8$	Crystals (hydrothermally crystallized, slightly impure)	4 days	990	Ga-orthoclase Xls
		3 days	1,001	Mostly Ga-orthoclase+some lower-index material, indicates beginning of incongruent melting
		24 hr.	1,010	Ga-orthoclase, glass, and Ga-leucite (not equilibrium)
		24 hr.	1,019	Ga-leucite and glass+sparse Ga-orthoclase (not equilibrium)
		24 hr.	1,036	Ga-leucite and glass
$KAlGe_2O_8$	Glass	24 hr.	1,111	Ge-orthoclase, rare interstitial glass
		3 hr.	1,115	Ge-orthoclase, sparse glass
		3 hr.	1,119	Ge-orthoclase in glass (50–50)
		23 hr.	1,123	All glass
$KGaGe_2O_8$	Glass	3 hr.	950	Ga-Ge-orthoclase+5 per cent small high-index needle-like Xls
		24 hr.	975	Ga-Ge-orthoclase+two high-index phases (sparse), needle-like and blocky Xls; very rare glass?
		23 hr.	985	As above+sparse interstitial glass
	Crystals	24 hr.	1,000	Glass, sparse Xls of Ga-Ge-orthoclase, + above two "impurity phases"

becomes smaller in the same way as that observed in the soda compounds, the order being normal orthoclase, Ga-orthoclase, Ge-orthoclase, Ga-Ge-orthoclase.

The melting points of the potash compounds, including normal orthoclase, are listed below. Thermal data are summarized in table 5.

$KAlSi_2O_8$ —1,170° C. (melts incongruently, to leucite and liquid)
$KGaSi_2O_8$ —1,000°–1,020° (melts incongruently, to gallium leucite and liquid)
$KAlGe_2O_8$ —1,122°
$KGaGe_2O_8$—approximately 1,000°

The incongruent melting of potash feldspar to leucite ($KAlSi_2O_6$) and a siliceous liquid is well known (Morey and Bowen, 1922). The behavior of Ga-orthoclase is analogous, melting to the Ga-containing equivalent of leucite and siliceous liquid. The new compound, "gallium leucite" ($KGaSi_2O_6$), has not been isolated from the glass or synthesized in pure form but was identified on the basis of the virtual identity of its X-ray powder pattern to that of natural leucite and on optical similarities. The close correspondence of the X-ray patterns indicates that Ga-leucite at room temperature has the same structure as low leucite. It is thus probable that it undergoes a polymorphic transition analogous to that in normal leucite. The upper temperature limit of existence of Ga-leucite in the melt (liquidus temperature) was not determined.

Ge-orthoclase and Ga-Ge-orthoclase

melt congruently. The reason for the difference in behavior is probably not due to inherent crystal-chemical differences between ordinary orthoclase and Ga-orthoclase, on the one hand, and Ge-orthoclase and Ga-orthoclase, on the other. Perhaps "germanium leucite" does not exist, or it may have a field of stability small enough that it does not impinge on that of the germanium orthoclases, thus permitting them to melt congruently. No attempt has been made to determine experimentally whether or not $KAlGe_2O_6$ is stable, but it is obvious that it does not exist under the composition and temperature conditions present at the melting points of Ge-orthoclase and Ga-Ge-orthoclase. It is the writer's impression that incongruent melting is considered by many to be a fundamental "property" of the compound exhibiting it. This melting behavior must, however, be viewed in terms of the equilibrium conditions within the system of elements present at the melting point of the compound. If incongruent melting occurs, it is because, at temperatures at and above the breakdown point, another phase assemblage is stable. Leucite and Ga-leucite, for example, are stable in a siliceous liquid above the temperature of breakdown of orthoclase and Ga-orthoclase. If, however, no compound of K_2O, Al_2O_3, Ga_2O_3, and GeO_2 is stable above the melting point of Ge-orthoclase or Ga-Ge-orthoclase, then these compounds *must* melt congruently. Thus incongruent melting is in a sense not a specific property of a compound but is dependent upon the stability of another phase or phases at higher temperatures.

The difficulties encountered with volatilization and high viscosity of the siliceous melts in the case of the albites are more pronounced with the potash compounds. Ga-orthoclase melt is exceedingly viscous and, in addition, quite refractory because of the persistence of Ga-leucite at high temperatures. Loss of K_2O is difficult to avoid. A precise determination of the incongruent melting point is difficult. Hydrothermal crystallizations again had to be resorted to, and the crystals thus obtained for melting determinations also had some foreign phase or phases present. Replacement of SiO_2 by GeO_2 results in a great decrease in viscosity of the melt, as in the case of the albites. Ge-orthoclase and Ga-Ge-orthoclase were readily crystallized from their glasses. The fairly large melting interval observed in Ga-Ge-orthoclase is probably a result of volatile loss of K_2O and/or GeO_2, with consequent composition change. The presence, as noted in table 5, of two foreign crystalline phases as inclusions also indicates that the composition is not precisely $KGaGe_3O_8$. The chemistry of Ga and Ge compounds is not well known, and little time has been spent in attempting to identify these excess phases. Chemical analyses of mixtures such as this are being made, however, and for future work the composition will be adjusted.

The lower melting point of the Ga as compared to the Ge compound is again noted. The effect is greater than in the soda compounds, although complicated by incongruent melting in Ga-orthoclase.

LIME FELDSPARS WITH Ga AND Ge

THE FULLY SUBSTITUTED COMPOUNDS

The Ga and Ge analogues of anorthite ($CaAl_2Si_2O_8$) can be crystallized,[7] but complete substitution of Ga for Al, Ge for Si, or both, apparently results in an

[7] After this manuscript was completed, it was found that Ga-anorthite had been earlier synthesized by Trömel in an investigation of the structural relations of anorthite and nepheline (Eitel, Herlinger and Trömel, 1930).

unstable or metastable structure. To produce stable structures, partial substitutions of Ga and Ge were resorted to, and this series of lime compounds possesses properties that make them the most interesting of the three groups of feldspars.

The fully substituted feldspars equivalent to anorthite are:

$CaGa_2Si_2O_8$ —"gallium anorthite"
$CaAl_2Ge_2O_8$ —"germanium anorthite"
$CaGa_2Ge_2O_8$—"gallium-germanium anorthite"

Melts of these compositions are quite fluid, and crystallization of the glasses takes place readily.

Homogeneous crystallizations of these compounds could not be produced; one or more additional phases[8] appeared in all preparations. Although some difficulty in obtaining precise composition was encountered in the case of Ge-anorthite because of GeO_2 volatilization in the rather refractory mixture, the extraneous phases were not due to the glasses being off-composition. The amount of nonfeldspar material varied, depending upon the temperature and, in some cases, upon the length of time of crystallization. In general, higher temperatures produced a lower "yield" of the feldspar, as did increasing the time of the run. Additions of small amounts of LiF as a flux also increased the amount of "foreign" material. Reacting the oxides directly in the solid state for the Ge-anorthite composition tended to produce less anorthite than was formed from the glass, although completion of the reaction is uncertain here. Melting points of the fully substituted lime compounds cannot be determined. Breakdown to a liquid plus the nonfeldspar phases occurs over a large temperature interval. This behavior is probably not due to incongruent melting, as the feldspars tend to persist over a range of temperature along with the other phases.

The observed thermal behavior of Ga-anorthite, Ge-anorthite, and Ga-Ge-anorthite seems to indicate that these compounds are not stable under the P and T conditions investigated. The "foreign" or "extraneous," etc., phases referred to are very probably those stable under the conditions of the experiments, the anorthites having formed as labile structures. Table 6 contains some of the pertinent thermal data in more detail than those presented above. The anorthite-containing runs listed in table 6 are not representative of equilibrium conditions. The liquidus temperatures of these compounds are here presented; the melting point of normal anorthite is included for comparison:

$CaAl_2Si_2O_8$	— 1,553° C.	(melting point)
$CaGa_2Si_2O_8$	— 1,323°	liquidus temperature
$CaAl_2Ge_2O_8$	—> 1,400°	liquidus temperature
$CaGa_2Ge_2O_8$	— 1,321°	liquidus temperature

The liquidus temperature of Ge-anorthite was not determined, but it is well above 1,400° C. Here again is seen the greater effect of Ga as compared to Ge in temperature lowering.

Although the anorthites appear to be unstable and cannot be crystallized free of some additional material, at certain temperatures these compounds can be obtained relatively pure. Ga-anorthite is best obtained at 900°, Ge-anorthite at 1,000°, and Ga-Ge-anorthite at 750° C. It was found that on the basis of optical homogeneity Ga-Ge-anorthite could be obtained in the purest form of the three, and Ge-anorthite the least pure. At any

[8] As in the case of the alkali feldspars, no attempt has been made to identify extraneous crystalline phases. To do so would involve a sizable expenditure of time, and it was not deemed necessary for the present investigation.

rate, it is possible to obtain X-ray powder patterns of these compounds without significant contamination of the pattern by the excess phases. In plate 2 the powder diffraction patterns of the lime compounds are shown, and the anorthite structure is apparent.

Table 7 lists the refractive indices of the fully substituted lime compounds, including normal anorthite. These crystals are, for the most part, highly twinned.

The tendency for a decrease in the gap between refractive indices of crystals and corresponding glasses has already been noted in the alkali feldspars, the interval being greatest in the normal feldspar and growing smaller in the order Ga compound, Ge compound, Ga-Ge compound. Table 7 shows the relations in the lime feldspars. The low (α) index of pure normal anorthite is virtually the same as that of anorthite glass. In Ga-anorthite the index of the glass becomes

TABLE 6

THERMAL DATA FOR THE LIME FELDSPARS

Composition	Original Condition	Time	Temp. (° C.)	Final Condition
$CaGa_2Si_2O_8$	Glass	25 hr.	850	Ga-An+fine-grained high n phase
		43 hr.	1,000	Ga-An+10 per cent or more high n phase
	Glass+LiF flux	43 hr.	1,000	Ga-An+>10 per cent high n phase
	Glass	5½ hr.	1,101	Ga-An+abundant lath and rod-shaped high n inclusions
		24 hr.	1,190	Glass (50 per cent)+at least 2 Xln phases—(1) tiny Xls, and larger rods and blocky Xls with high n, inclined extinction, moderate birefringence; (2) lesser amounts of Ga-An
		1½ hr.	1,220	Glass+blocky and needle-like high n prisms
		3 hr.	1,254	Glass+blocky Xls
		3¼ hr.	1,313	Glass+sparse tiny high n Xls
	Xls in glass	2¼ hr.	1,321	Glass+rare tiny Xls
		3 hr.	1,325	All glass
$CaAl_2Ge_2O_8$	Glass	25 hr.	850	Inhomogeneous Xln aggregate; fine "perthitic-like" intergrowth of at least two phases
		18 hr.	1,000	"Striated" Xls of Ge-An (at least one other phase)
	Glass+LiF flux	7 days	1,000	Heavily "striated" Xls of Ge-An, full of high n inclusions and very cloudy; Ge-An content much lower than above material
	Glass	22 hr.	1,200	Ge-An Xls, full of aligned high n inclusions
	Oxides	24 hr.	1,200	Xls Ge-An, full of greenish, large, irregular high n, low biref. Xls (>50 per cent?)
	Oxides+LiF	24 hr.	1,200	At least two isotropic high n Xln phases
	Glass	30 min.	1,300	Inhomogeneous Xln material
		2¼ hr.	1,300	Glass (50 per cent), Ge-An Xls, blocky high n Xls
		30 min.	1,400	High n Xls in glass
$CaGa_2Ge_2O_8$	Glass	4 days	750	Ga-Ge-An+5 per cent of a high n material
		24 hr.	900	Ga-Ge-An+moderate amounts of high n material
		5½ days	900	Ga-Ge-An+over 50 per cent high n material
		3 hr.	1,204	Glass+two Xln phases (one may be Ga-Ge-An)
		3 hr.	1,254	As above, more glass
		3 hr.	1,296	Glass+a high n (orthorhombic?) phase
		2½ hr.	1,321	Very rare Xls in glass
	Xls	2½ hr.	1,321	All glass

slightly greater than the high index of the crystalline form. In Ge-anorthite this difference is more pronounced, and in Ga-Ge-anorthite the crystals have indices very much lower than the glass of their own composition.

The densities of these compounds and their glasses have not been determined, and it is doubtful that density determinations on these finely crystallized and impure substances would be good enough to be of much value. The fact that the refractive indices of the glasses are greater than those of the crystals might indicate similar density relations. It would be of interest to know the true density relations between these crystals and glasses in connection with the problem of the instability of the crystalline phases. It would be exceedingly difficult, however, to obtain these values at the elevated temperatures of equilibrium, and the relative relationships at room temperature might change at the temperatures of crystallization. The reversal of the normal crystal-glass indices observed might, however, be an indication of instability of the crystalline modifications.

Very few silicates have densities less than their corresponding glasses. The *Handbook of Physical Constants* (Birch, Schairer, and Spicer, 1942) lists but two: pure synthetic akermanite ($Ca_2MgSi_2O_7$) and $K_2Si_4O_9$, which is not known to exist as a mineral. The density of pure akermanite is 2.944, its glass is 2.955, a difference of 0.4 per cent. Crystalline $K_2Si_4O_9$ has a density of 2.335; its glass is 2.384, 0.2 per cent greater. Pure akermanite is, however, quite rare, if it exists at all in nature, and again the relative densities at the melting temperatures are not known. The small difference in refractive indices of crystalline anorthite and anorthite glass is expressed by a small difference in density, 2.765 for the crystals and 2.700 for the glass. This is but 2.35 per cent, whereas albite crystals and glass differ by 8.6 per cent. In the light of the index data and labile nature of the fully substituted Ga- and Ge-anorthites, it might be that pure anorthite comes close to being an unstable mineral.

TABLE 7

REFRACTIVE INDICES OF FULLY SUBSTITUTED LIME COMPOUNDS AND THEIR GLASSES

Refractive Index	$CaAl_2Si_2O_8$	$CaGa_2Si_2O_8$	$CaAl_2Ge_2O_8$	$CaGa_2Ge_2O_8$
Glass	1.5755	1.633	1.658	1.745
Crystals				
α	1.576	1.625	1.641	1.705
γ	1.588	1.631	1.647	1.711

PARTIAL SUBSTITUTIONS OF Ga AND Ge IN ANORTHITE

To determine how much Ga or Ge the anorthite structure can take and remain stable, two compositions were prepared; one in which half the Al was replaced by Ga and one in which half the Si was replaced by Ge. The thermal behavior of these compositions is quite interesting, and, in order to evaluate this behavior more fully, anorthites with one-quarter and with one-eighth of the Al replaced by Ga were synthesized. These compositions are represented and designated as follows:

$CaGaAlSi_2O_8$ —"½ gallium anorthite"
$CaAl_2SiGeO_8$ —"½ germanium anorthite"
$Ca_2(GaAl_3)Si_4O_{16}$ —"¼ gallium anorthite"
$Ca_4(GaAl_7)Si_8O_{32}$—"⅛ gallium anorthite"

X-ray powder photographs of the half-substituted crystals are included in plate 2.

The thermal data are presented in table 8. These compounds are stable, but it is not yet known how much more than

half the Si or Al can be replaced before they become unstable. The melting points of the partially substituted anorthites are as follows:

$Ca_4(GaAl_7)Si_8O_{32}$—1,523° C.
$Ca_2(GaAl_3)Si_4O_{16}$—1,488°
$CaGaAlSi_2O_8$ —1,428°
$CaAl_2SiGeO_8$ —1,465°

These are well-defined melting points; a small interval is observed in the case of ½ Ge-anorthite, but it is probably due to the mixture's being somewhat off-composition. An excess phase, which has the optical characteristics of β-Al_2O_3, is found in runs of this composition and is probably present as a consequence of GeO_2-loss during fusion of the mix. It is again observed that the Ga compound has a lower melting point than the Ge equivalent.

The sharp melting points observed in the half-substituted compounds were unexpected. Consider only the Ga-anorthites, neglecting the Ge compounds, which, for the purposes of discussion, are equivalent. $CaGaAlSi_2O_8$ can be viewed as a solid solution, midway between anorthite and Ga-anorthite in composition. Even though the gallium end-member is unstable, it might be expected that the stable mid-point would exhibit a melting interval that is typical of solid solutions. The ¼ and ⅛ compounds were prepared in order to evaluate this situation; for, if the ½ compound represented a composition at or near the minimum temperature on a solid-solution curve with a minimum, it would melt over little or no temperature interval. The ¼ and ⅛ Ga-anorthites were also found to melt sharply, however, within the limits of experimental error. Thus all these compositions (and also ½ Ge-anorthite) have a melting behavior characteristic of pure "end-member" compounds and unlike that of mix crystals.

TABLE 8

THERMAL DATA FOR THE PARTIALLY SUBSTITUTED LIME COMPOUNDS

Composition	Original Condition	Time	Temp. (° C.)	Final Condition
$CaGaAlSi_2O_8$	Glass	1 hr.	1,426	Xln ¼ Ga-An
		1¼ hr.	1,430	All glass
$Ca_2(GaAl_3)Si_4O_{16}$	Glass	30 min.	1,486	Xln ¼ Ga-An
	Crystals	30 min.	1,490	All glass
	Glass	4 hr.	1,487	Xln ¼ Ga-An
	Crystals	4 hr.	1,487	Xln ¼ Ga-An
	Glass	5¼ hr.	1,495	All glass
	Crystals	5¼ hr.	1,495	All glass
$Ca_4(GaAl_7)Si_8O_{32}$	Crystals	30 min.	1,516	Xln ¼ Ga-An
		30 min.	1,521	Xls ¼ Ga-An in glass (>½ glass)
	Glass	30 min.	1,525	All glass
	Crystals	4 hr.	1,517	Xls ¼ Ga-An
	Glass	4 hr.	1,517	Xls ¼ Ga-An
	Crystals	5½ hr.	1,525	All glass
	Glass	5½ hr.	1,525	All glass
$CaAl_2SiGeO_8$	Glass	1¾ hr.	1,457	Xln ½ Ge-An+small amounts of β-Al_2O_3 (?)
		1 hr.	1,461	Xln ½ Ge-An+glass(½)+some β-Al_2O_3 (?)
		1½ hr.	1,465	Glass+some β-Al_2O_3 (?)+several small Xls of ¼ Ge-An

A melting diagram of the system $CaAl_2Si_2O_8$-$CaGa_2Si_2O_8$ is presented in figure 1. Additional compositions are necessary to complete the diagram, particularly in the region beyond ½ Ga-anorthite, where the stability conditions are unknown. The stable portion of this system appears to be unique, however; the writer knows of no other systems in which a solid solution exists that can be represented by a single straight line and without distinct solidus and liquidus curves.

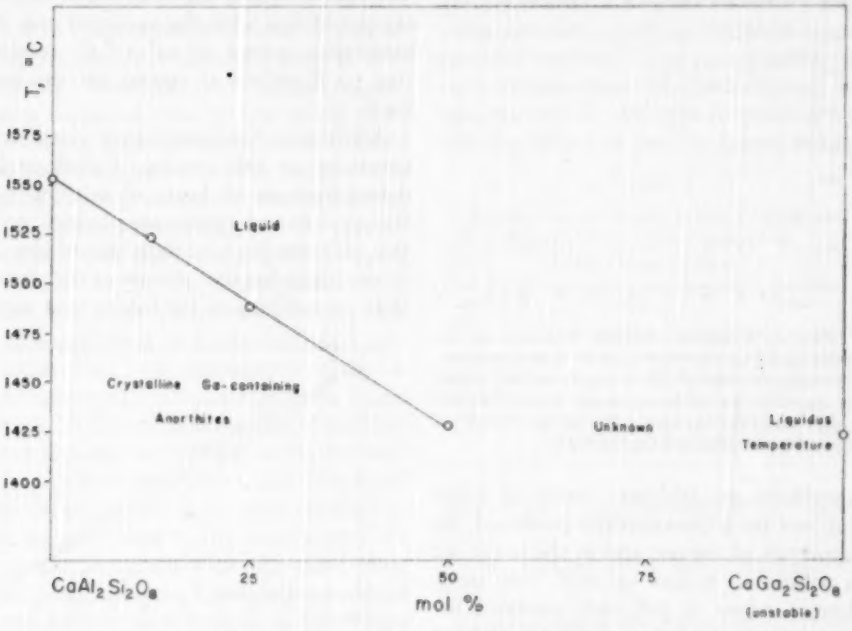


FIG. 1.—Melting diagram of the system $CaAl_2Si_2O_8$-$CaGa_2Si_2O_8$

The observed relations shown in figure 1 are puzzling. The single line probably represents the very close approach of the normal liquidus and solidus curves, the melting interval being so small that it cannot be detected within the experimental error of temperature and composition control. Bowen (1913), in his well-known paper on the plagioclase feldspars, assumed perfect solutions and thus derived equations for the liquidus and solidus curves in a complete series of solid solutions. The calculated curves fit his experimental data very well indeed. He calculated the heat of melting of anorthite to be 29,000 cal/mol (104 cal/gm). Seltz (1934), using fugacities and activities, derived similar expressions. These relations are such that the liquidus and solidus curves approach each other as the latent heat of melting of either end-member becomes very small, and a single straight line can result only if the two end-members melt at the same temperature. If, however, the latent heats of the end-members equal or are close to each other ($\Delta H_1 \simeq \Delta H_2$), the liquidus and solidus curves tend to become symmetrical about a straight line; and if both are

small, a straight-line relation is approached.

Figures 2 and 3 are computed melting diagrams in which the latent heats have been varied. The composition of ½ Ga-anorthite has been used as an end-member merely to facilitate computation and should not be considered as the real end-member compound. In figure 2 Bowen's value of 29,000 cal/mol is used as ΔH for

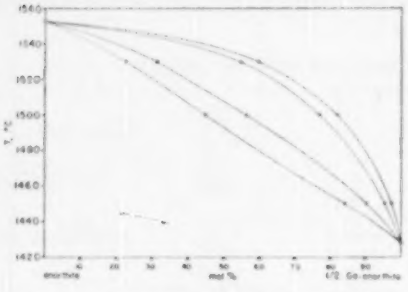


FIG. 2.—Computed melting diagrams of anorthite and ½ Ga-anorthite (used as an end-member). The accepted value of ΔH = 29,000 cal/mol is used for anorthite, and arbitrary values of 5,000 cal/mol (upper set of curves) and 20,000 cal/mol (lower set of curves) are used for ½ Ga-anorthite.

anorthite; an arbitrary value of 5,000 cal/mol for ½ Ga-anorthite produced the upper set of curves, and in the lower set a value of 20,000 cal/mol was used. Lower values of ΔH were assumed for anorthite in figure 3: 20,000 cal/mol for both latent heats produced the wider set of curves, and 5,000 cal/mol for both end-members produced the narrower set.

These diagrams illustrate the problem at hand. The experimentally determined curve is not easily reconciled with the large value of ΔH for anorthite. Assignment of a very small latent heat to ½ Ga-anorthite would produce a very small melting interval, but the melting curves would be nonlinear. One might expect just this relation to hold, as it is quite possible that the heat of melting of the Ga-containing anorthites becomes very small; their unstable nature beyond a certain Ga concentration would suggest this. The narrow loop of figure 3 matches the experimental data very well. The maximum spread in the loop is but 3°–4° C., which, if determined experimentally, would be plotted as a straight line. The value of ΔH used for anorthite in this calculation (5,000 cal/mol) is, however, far out of line with the accepted one. A maximum spread of 11°–12° C. results (fig. 3) if values of 20,000 cal/gm are used.

Additional thermodynamic data are necessary in this system. Calorimetric determinations of heats of solution on the crystals and glasses are planned, and this plus specific heat data should give a more comprehensive picture of the situation. In addition to the latent heat data

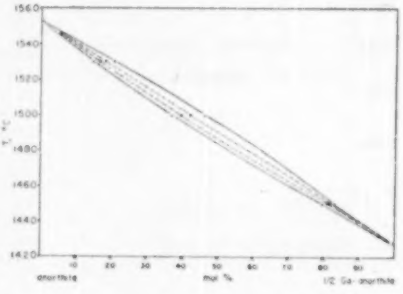


FIG. 3.—Same as fig. 2 (computed curves) except that arbitrarily lower values are also used for anorthite. Latent heats of 20,000 cal/mol used for both end-members in the wider set of curves, and 5,000 cal/mol for both in the narrower (dashed) set.

there are several other factors that should be considered and that might be effective in producing the observed straight-line relation:

a) These mixtures may be nonideal in character. The first addition of Ga may be the most effective in lowering the latent heat of fusion of the mix-crystals,

i.e., the ΔH curve for the mixtures may be nonlinear.

b) The composition of $\frac{1}{2}$ Ga-anorthite has arbitrarily been used as an end-member in these calculations. It is not yet known how far beyond this composition the anorthites are stable, but consideration of an end-member richer in Ga would affect the results.

c) If diffusion of Ga and Al is very slow in the process of melting or crystallization, the crystals might tend to melt sharply. Assuming that under equilibrium conditions the system would show a melting loop, lack of diffusion would prevent enrichment in the liquid of the lower melting component during melting (and prevent enrichment in the crystals of the higher melting component during crystallization). The mix-crystals would then behave as a one-component system and would melt to a liquid of their own composition. An apparently straight-line nonequilibrium melting curve might result. Thirty-minute runs had been used to determine the melting curve, but several 4–5-hour runs were made on $\frac{1}{8}$ and $\frac{1}{4}$ Ga-anorthite to check this possibility. As seen in table 8, the results on both crystals and glass were the same as in the 30-minute runs. Considering the high temperatures involved, it is not likely that diffusion would be slow enough to prevent equilibrium melting or crystallization. It is also to be noted that this effect is not observed in the albite-anorthite system, where crystal-liquid equilibrium involves diffusion not only of Al and Si but also of Na and Ca.

The question of the "degree of perfection" of the Ga and Ge substitutions in anorthite is of interest in connection with the stability relations of pure anorthite. Al and Si can be completely replaced by Ga and Ge in the alkali feldspars, and the structure remains stable. The fact that the lime feldspar will not accept complete replacements of Al and Si must be due to a difference between anorthite and the alkali feldspars, as the substitutions are the same. If the substitution is perfect or near-perfect in the alkali feldspars, why do anorthites become unstable when something over half the Al or Si is replaced by Ga or Ge? It was previously stated that pure anorthite may come close to being an unstable compound. Complete Ga or Ge replacement, even though these substitutions may be near-perfect, is sufficient to produce instability; this fact may be a good indication of the "near-instability" of anorthite.

TABLE 9

REFRACTIVE INDICES OF THE PARTIALLY SUBSTITUTED LIME COMPOUNDS AND THEIR GLASSES

Refractive Index	$Ca_2(GaAl_3)Si_4O_{16}$	$CaGaAlSi_2O_8$	$CaAl_2SiGeO_8$
Glass	1.591	1.608	1.611
Crystals			
α	1.591	1.604	1.608
γ	1.596	1.611	1.615

The optical data are presented in table 9.

Comparison of the glass-crystal refractive-index relations of these stable compositions with the fully substituted compounds is interesting. Both of the half-substituted compounds have glass indices that are approximately the mean of the crystal indices. The $\frac{1}{4}$ Ga compound has relative values similar to ordinary anorthite. The fact that the half-substituted compounds have average refractive indices equal to their glasses appears to be in line with the above discussion of stabilities. This might indicate a very small value of ΔH for these compositions, in which case the free energies

of the liquid and crystals would be nearly the same.

THE RELATIVE EFFECTS OF Ga AND Ge ON THERMAL PROPERTIES

In all the compounds considered it has been observed that Ga has a greater influence on the melting temperatures than does Ge. The weakening of the structures by Ge replacement is not very great, as observed from the melting points of the Ge compounds as compared to the normal feldspars. In the alkali feldspars the Al-Si ratio is 1:3, yet the one-atom Ga replacement is much more effective in lowering melting temperatures than is the three-atom Ge replacement. The liquidus temperatures of the unstable anorthites show this same relation; and Ga-anorthite has approximately the same liquidus temperature as does Ga-Ge-anorthite.

The reasons for this behavior, particularly in complex crystals of the feldspar type and in liquids composed of three or four oxides, are not easily evaluated; the whole crystal-liquid energy picture is not known, and the computation of bond strengths is exceedingly difficult and would be of doubtful significance. The melting phenomena of silicates in general as related to structure is complex and has not been successfully treated. Wickman (1943) briefly considers the problem and points out the great thermal stability of network-type silicates with a 1:1 Al-Si ratio. Buerger (1948), in discussing the melting relations with respect to the number of shared oxygen atoms (in network structures, such as the feldspars, all oxygen atoms are shared), states:

> Consider now the effect of aluminum. If the aluminum proxies for silicon in certain tetrahedra, the saturation of the oxygens of those tetrahedra is reduced and they are capable of contributing to stronger bonding between silicate units than without the aluminum. In this way, the presence of aluminum proxying for silicon in a silicate increases its disintegration temperature and consequently raises its position in the reaction series. This is true for both discontinuous and continuous reaction series. The effect of aluminum in a discontinuous reaction pair can be seen in the higher position of leucite (Al:Si = 1:2) with respect to orthoclase (Al:Si = 1:3). The effect of aluminum in a continuous reaction series is illustrated by the higher position of anorthite (Al:Si = 1:1) with respect to albite (Al:Si = 1:3).

The structural picture as it effects melting behavior is quite complex, and Buerger's discussion is an oversimplification. The effect of the cation is not considered; yet its bonding energy cannot be neglected. The melting points of the following anorthite-like compounds illustrate this:

$CaAl_2Si_2O_8$ (anorthite)	—1,553° C.
$SrAl_2Si_2O_8$	—1,660°
$BaAl_2Si_2O_8$	—1,720°

In the example of the anorthite-albite melting relations, consideration of only the Al-Si ratio and neglect of the cation change is perhaps not justified.

In orthosilicates, with isolated tetrahedra, melting can occur without breaking tetrahedral bonds. In structures with an increasing number of shared O atoms, an increasing number of tetrahedral bonds may be broken during thermal breakdown. In feldspar-type structures, where sharing of O is complete, these bonds *must* be broken, and many of them must be disrupted in order to melt the crystal. Not neglecting the cations, it is difficult to say a priori where the weakest points for breakdown will be. The relatively low melting behavior of the Ga feldspars might indicate, however, that, irrespective of the strengthening effect of a trivalent atom (and its attendant divalent cation) in a network

alumino-silicate, the weak link is a bond to the trivalent atom.

Another explanation that is not dependent on an analogy between Ga and Al in the structure might account for the large effect of Ga on the melting points. Dr. Fritz Laves has pointed out to the writer that, in many intermetallic compounds, Ga is highly polarized. If the Ga atom is distorted in the feldspars, its bond strengths to the surrounding four O atoms may be nonuniformly distributed; it may be preferentially bonded to one, two, or three oxygens. If so, the correspondingly weakened bond or bonds may be responsible for breakdown at lower temperatures.

SUMMARY AND CONCLUSIONS

The synthesis of feldspars in which Al and Si have been replaced by Ga and Ge is described, and some of their crystal-chemical properties described. One of the principal reasons for the synthesis of these compounds is the better evaluation of the order-disorder relations within this group of minerals; the present paper does not, however, consider this phase of the investigation. The work here described is but a beginning, and it is hoped that additional work with these synthetic compounds will lead to a better understanding of the natural alumino-silicates. In addition to making use of the difference in X-ray scattering power produced by Ga or Ge substitution, the somewhat "weaker" models produced might more readily exhibit some of the structural changes that are so hard to induce in certain of the alumino-silicates.

With the exception of investigating a portion of the incomplete solid-solution series, anorthite–gallium anorthite (and the 50 per cent Ge substitution in anorthite), only the "end-member" feldspars have been synthesized. Synthesis of the intermediate Ga and Ge plagioclase compositions has not yet been attempted, nor has synthesis of the potash-soda solid-solution series. In the light of the instability of the fully substituted Ga- and Ge-anorthites, it will be of interest to determine the relation between the Na-Ca ratio and the amount of Ga and Ge that is accepted in the plagioclases. The use of partial instead of complete substitutions of Ga and Ge in the alkali feldspars may also prove of value in the interpretation of these structures.

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THE OPTICS OF TRICLINIC ADULARIA[1]

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ABSTRACT

Adularia traditionally has been described as monoclinic. Sections of adularia crystals from several localities, studied on the universal stage, show optical orientations inconsistent with monoclinic symmetry. The optical deviation from monoclinic symmetry is absent or very small in the cores of the crystals but increases toward the surfaces. The optical orientation and axial angles of triclinic adularia vary from those of monoclinic adularia in the direction of sanidine rather than of microcline. Triclinic adularia occurs as a secondary modification of monoclinic adularia. It has distinctive optical behavior which establishes it as a variety of potash feldspar.

INTRODUCTION

Former investigators have described four optically distinct types of potash feldspar: adularia, orthoclase, sanidine, and microcline. Adularia, orthoclase, and sanidine traditionally have been described as monoclinic; microcline is triclinic. The name "orthoclase" has both a general and a more specific usage. It is occasionally applied to the whole family of monoclinic potash feldspars, occasionally to a subgroup within that family. This subgroup is poorly defined. In much of the literature the name "orthoclase" is commonly used for any monoclinic potash feldspar which does not possess the distinctive characteristics of adularia or sanidine. And even these two minerals are defined differently by different writers. In this paper the general relationships between the potash feldspars as reported in the literature are outlined, some recognized complications are reviewed, and further complications introduced by the present findings are described and evaluated.

The orthoclase family consists of three members: adularia, ordinary orthoclase, and sanidine. All three are commonly said to be structurally and optically monoclinic, with (010) as the plane of symmetry. Adularia is generally considered to be the low-temperature member, usually restricted in occurrence to vein deposits and ore bodies. It has a distinctive crystal habit limited almost entirely to the forms (110), (001), and (T01). In most descriptions it is characterized optically by extinction parallel to (010) in the zone [010]; by the position of the optic plane, which is normal to (010) and nearly parallel to (001); and by a negative axial angle of about 50°–70°, a being, in all cases, near [100].

Ordinary orthoclase is not distinguished by any single mode of occurrence or crystal habit, as is adularia. It is optically similar to adularia, although some writers, such as Spencer (1937), report that "true" orthoclase has a smaller average $2V$ (25°–50°) than has adularia.

Sanidine, the high-temperature member of the orthoclase family, is usually found in dikes, stocks, flows, etc. It, too, is monoclinic, but optically it differs from both ordinary orthoclase and adularia. In sanidine the optic plane may be either normal or parallel to (010), and the negative axial angle is variable (0°–60°).

Perhaps the best optical classification of these feldspars was that given by

[1] Manuscript received May 20, 1950. Presented at the November, 1949, Annual Meeting of the Geological Society of America.

Spencer in his paper on the thermal stability of the potash-soda feldspars. His classification is outlined graphically in figure 1. Spencer distinguishes between the three members of the orthoclase family principally by the size of $2V_\alpha$. Sanidine, for example, has a $2V$ of less than 25° when the optic plane is normal to (010); but, if the optic plane is parallel to (010), $2V$ may be as large as 50°–60°. Thus in the orthoclase family the optic plane tends to be parallel to the symmetry plane if the temperature of crystallization is high, and to be normal to it if the temperature is low. Adularia and ordinary orthoclase have been subjected to heating experiments by a number of investigators, including Des Cloizeaux (1861, 1862), Merwin (1911), Kôzu (1925), Barth (1931), and Spencer (1937). In general, adularia resists optical change more stubbornly than does ordinary orthoclase, but there is considerable variation in behavior from specimen to specimen of either mineral. When held for periods of about 100–300 hours at temperatures varying between 900° and 1,120° C., the optic angles of most specimens decrease to 0° and open out again in the symmetry plane (010). Thus ordinary orthoclase and adularia assume sanidine optics upon proper heat treatment.

Microcline is triclinic. Its crystallographic angle, γ, deviates from 90° by 2½° (see accompanying paper by Laves in this issue of the *Journal*), but this small deviation is expressed optically by a striking extinction angle of 15°–20° measured from the (010) cleavage in (001). The optic plane is nearly perpendicular to the (010) cleavage. The optic angle is large, about 80°(−). Microcline undergoes some optical change upon heat treatment, but it is decidedly more resistant to change than is orthoclase. Some, but not all, specimens of microcline lose their cross-hatch structure and acquire sanidine optics after heating at about 1,075° C. for 300 hours or more (Spencer, 1937). Microcline and adularia are characteristically more resistant to sanidinization than is ordinary orthoclase.

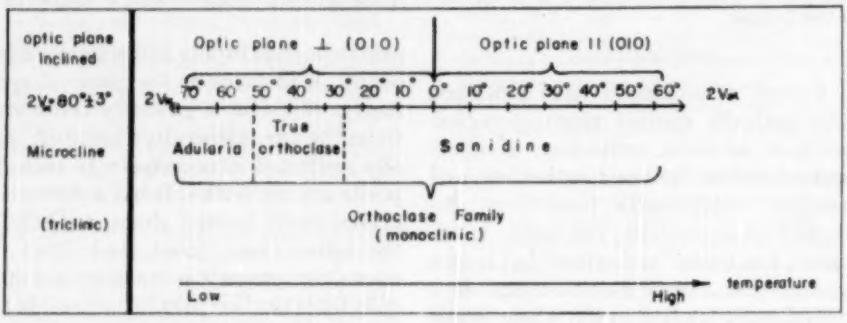

FIG. 1.—Outline of the optical classification of the potash feldspars modified from Spencer (1937)

The two stereograms of figure 2 illustrate the optical orientation of these four types of potash feldspar as seen on a stereographic projection normal to the *c*-axis.

EARLIER INTERPRETATIONS

The stereograms of figure 2 and the foregoing description present the classical

picture of the optical relationships among the potash feldspars. It is an oversimplified picture, and numerous observed deviations from it have long been a source of concern among mineralogists. Some believe that the generally monoclinic optics of the orthoclases and the triclinic optics of microcline express two fundamentally different crystal lattices. They embellish their argument by pointing out that the two forms have slightly

consideration of this paper. Mallard's conclusion has had many supporters, including Michel-Lévy (1879), Groth (1889), Hintze (1893), Brögger (1890), and Barth (1929). Because his optical observations presented a problem which to this day remains in dispute, Mallard's original description of the optics of triclinic adularia is given here.

Mallard obtained small, clear crystals of adularia from the vein deposits of St.

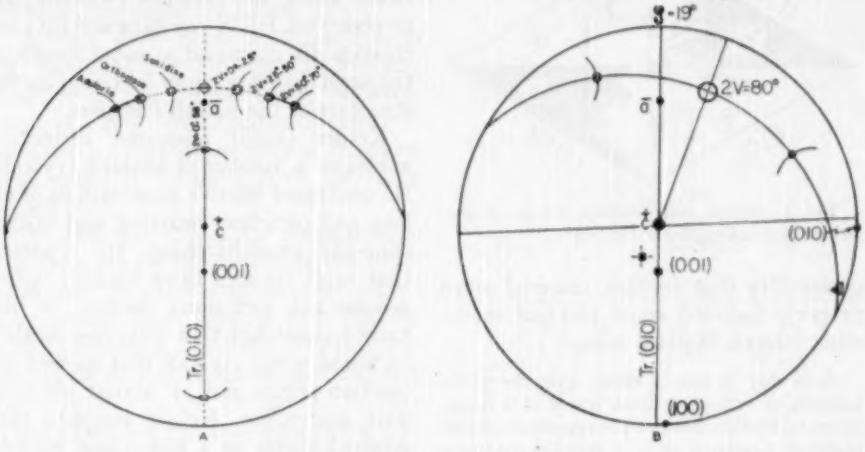

FIG. 2.—Stereograms illustrating the optical orientations of *A*, the orthoclase family, and *B*, microcline

different indices of refraction, specific gravities, and behaviors upon heating (see, e.g., Winchell, 1933, p. 354). That these differences are great enough to be significant is denied by others.

Other workers believe that there is structurally only one potash feldspar; that it is triclinic; and that what appear to be monoclinic optical properties are the illusion created by submicroscopic twinning of triclinic individuals. In 1876 Mallard studied the optical results of twinning in a number of minerals, including orthoclase and microcline, and first observed the peculiar triclinic optical effects in adularia which are the primary

Gotthard, Switzerland. He made thin sections normal to the *c*-axis and studied them under the polarizing microscope. In this section the crystals have a rhombic outline, the short diagonal of which is parallel to the (010) cleavage. Mallard noted that the rhombs did not behave as single crystalline units but that the centers of the rhombs showed extinction parallel to (010); and the edges showed extinction angles of 2°–3° from (010). The edges of the rhombs behaved optically as though they were divided into four sectors, as illustrated in figure 3, which is sketched from Mallard's publication. The (110) and ($\bar{1}$10) areas went to ex-

tinction simultaneously in the positive direction; ($\bar{1}10$) and ($1\bar{1}0$), simultaneously in the negative direction. Thus, although the cores of the adularia crystals appeared to have monoclinic extinction, the edges were optically triclinic. Mallard did not believe that a truly monoclinic mineral could produce triclinic optical effects, but he analyzed carefully the

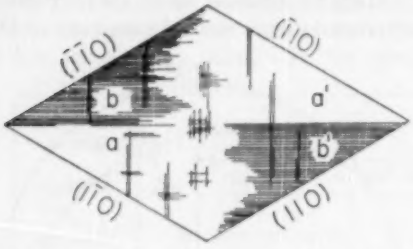

FIG. 3.—Sketch, after Mallard (1876), of adularia crystal cut normal to the *c*-axis.

probability that triclinic material when properly twinned could produce monoclinic effects. Mallard stated:

> It is easy to see, in effect, that the purest samples of orthoclase show traces of a fundamentally triclinic lattice . . . crystals of adularia from St. Gotthard show a true passage from microcline to theoretically homogeneous orthoclase. It is these passages which, to my mind, do not permit the separation of orthoclase and microcline into different species [author's translation].

Such a conclusion could be neither substantiated nor disproved by optical data alone. Numerous mineralogists have used optical, X-ray, and thermal methods to study the general problem of the potash feldspars; but, since Mallard, apparently only three people have investigated the specific problem of triclinic adularia.

Federov, in 1912, commented that etch figures on the surfaces of adularia crystals from St. Gotthard indicated some deviation from monoclinic symmetry. He made no optical study of the crystals.

Barth (1928, 1929) studied thin sections of adularia and described microcline-like cross-hatching. He reported albite, pericline, and acline twinning. All these are triclinic twins. However, because Barth reports that his X-ray studies show the lattice of this adularia to be only 10′ from monoclinic symmetry, it is difficult to see how any difference between albite and pericline twinning can be observed. His X-ray data and his conclusions are discussed at some length in the accompanying paper by Laves on the structure of the potash feldspars.

Köhler (1948) measured extinction angles on a number of adularia crystals. He confirmed Barth's observations of albite and pericline twinning and microcline-like cross-hatching. He reported that most crystals show triclinic optics around any inclusions, fissures, or disturbed zones but that from one locality he found whole crystals that showed extinction angles and $2V$ almost identical with microcline. Köhler suggests that adularia forms as a monoclinic mineral and then changes to microcline. The change begins in a patchy manner at the edges or around any disturbed areas within the crystal and eventually spreads over the entire crystal. Thus Köhler believes that adularia is a triclinic mineral, but he is not satisfied that his optical observations are sufficient for the drawing of final conclusions.

METHOD OF THE PRESENT INVESTIGATION

Former optical work on adularia has been limited almost entirely to studies of extinction angles on oriented thin sections. The present work is an attempt to correlate detailed universal-stage determinations with X-ray studies.

Adularia crystals from several widely

separated localities were obtained, and at least two, or even three or four, differently oriented thin sections were made of each crystal. Optical measurements were made at several spots on each section, after which the section was removed from its mounting and X-ray precession, rotation, or Laue pictures were made of the precise spots measured on the universal stage. The optical data will be discussed here, the X-ray data in the accompanying paper by Laves.

Studies were made of adularia from St. Gotthard and Tavetschtal, Switzerland; Mount Vesuvius, Italy; Salzburg, Austria; and Guanajuato, Mexico. A striking consistency in optical behavior was noted in adularia from all these localities.

At each spot chosen for measurement the three axes of the indicatrix, the optic axes, and the poles of all cleavage planes and crystal faces present were located and plotted on a Wulff net. The position of the *c*-axis was deduced, plotted, and then rotated to the center of the net. Thus all optical and crystallographic elements, after rotation in the same direction and of the same amount as the *c*-axis, are projected on a plane normal to the *c*-axis. In all stereograms the trace of the plane (010) is the north-south diameter of the figure. This makes possible direct comparison of all the stereograms.

In monoclinic crystals the *a*-axis of the indicatrix emerges on the (010) cleavage trace. In triclinic crystals it deviates from the (010) plane by varying amounts. The plane including *a* and the *c*-axis rotates about the *c*-axis in the same direction as the plane containing the *a*- and *c*-axes of the lattice. This is proved by X-ray pictures of carefully oriented crystal fragments. The optical rotation is measured with reference to the following system of co-ordinates:

$(010)\wedge(001) = 89°50'$,
$(010)\wedge(100) = 89°33'$,
$\alpha = 89°57'$,
$\beta = 116°$,
$\gamma = 90°24'$.

On the stereograms $(010)\wedge ac$ is arbitrarily called ϕ. There is also, in the tri-

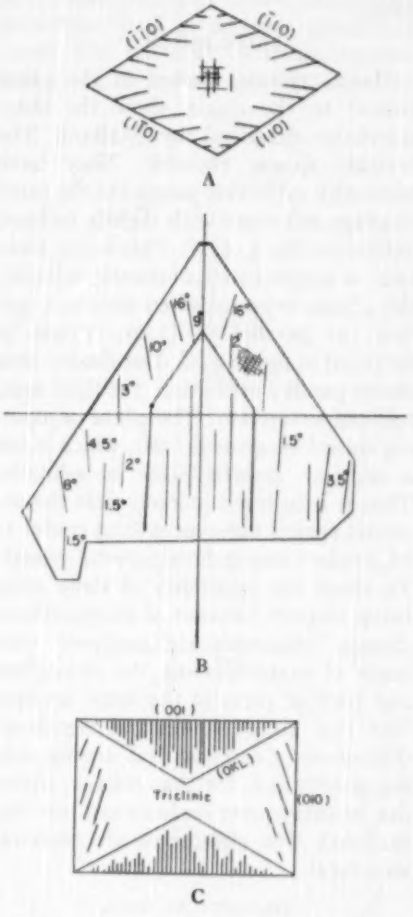

FIG. 4.—*A*, schematic drawing of adularia crystal cut normal to the *c*-axis, illustrating variation in extinction angles from core to edges. *B*, drawing of adularia crystal from St. Gotthard, showing measured values of extinction angles in section normal to the *c*-axis. *C*, schematic drawing of crystal from Mount Vesuvius, showing monoclinic and triclinic areas as seen in section cut parallel to (100).

clinic material, a clockwise (as seen in the positive direction of the *a*-axis) rotation of the optic plane about the *a*-axis of the indicatrix. The size of ϕ and the amount of the rotation of the optic plane are more or less independent measures of the deviation from monoclinic symmetry.

FALSE ZONING

Many crystals, viewed in the plane normal to the *c*-axis, show the characteristics described by Mallard. The crystals appear rhombic. They have cores with extinction parallel to the (010) cleavage and edges with slightly inclined extinction (fig. 4, *A*, *B*). This is not, however, a simple core-and-mantle relationship. Some crystals, when seen in a section cut parallel to (100), appear as sketched in figure 4, *C*. The shaded area shows parallel extinction, the light area, inclined extinction. The plane separating these two areas is (0*kl*), which is not a common growth plane in adularia. Thus it is believed unlikely that the observed optical differences from center to edge reflect zoning during crystal growth. To check the possibility of these areas being distinct because of compositional change, spectrographic analyses[2] were made of material from the monoclinic and triclinic parts of the same crystals. The two areas showed no significant differences. Compositional zoning during growth and, for that matter, zoning due to subsequent replacement are thus excluded. The differences are therefore structural.

THE OPTICAL DATA

Several crystals of adularia from St. Gotthard, Switzerland, were measured. The optical behavior typical of the St. Gotthard material is illustrated in the stereograms of figure 5. In this figure, *A* shows the optics of a tiny, clear crystal which had grown upon a much larger one. It behaved as a single, wholly monoclinic crystalline unit and conformed to the classical picture of adularia.

Figure 5, *B* and *C*, represent the large crystal upon which the smaller one of figure 5, *A*, had grown. It shows radically different optical behavior. No part of the crystal is optically monoclinic. At the core the angle ϕ is 4°.5, $2V = 58°$ (−). The optic plane is slightly inclined from the normal to (010). At the edge of the crystal, ϕ is 15°.5, $2V = 47°$ (−). The optic plane is decidedly inclined from the normal to (010). Figure 5, *D*, illustrates the peculiar behavior of a crystal consisting of a pair of albite twins. The optical orientation of each individual is referred to the same above-mentioned geometrical system of co-ordinates. The X-ray pictures of the two individuals showed no structural differences. Nevertheless, the two optic planes are oriented differently. As adularia crystals lose their monoclinic symmetry, the optic planes depart from a plane normal to (010) and rotate in varying degrees around the *a*-axis of the indicatrix. In this twinned crystal, one optic plane has rotated clockwise, the other anticlockwise about *a*. This is the only crystal measured which showed an anticlockwise rotation of the optic plane.

Figures 6 and 7 illustrate the optical behavior of adularia from Mount Vesuvius, Italy, and Guanajuato, Mexico. Figure 6 shows the optical variations within a crystal typical of the material from Mount Vesuvius. The crystal is twinned after the albite law, with (100) the *composition* plane. This plane zigzags somewhat in the zone of the *c*-axis. The material on either side of the (100) plane is somewhat cloudy and shows

[2] Spectrographic analyses kindly made by O. Joensuu. The Mount Vesuvius crystals investigated had an average Na_2O content of 0.4 per cent.

faint traces of poorly defined lamellae too small to be measured as individuals. These may possibly be pericline twins. They give a good, easily measured, average extinction. The areas measured are indicated on the sketch of the crystal (fig. 8). Areas *2* and *3* are essentially identical albite twins, as are areas *4* and *5*. Therefore, both stereograms 6, *B* and *C*, are an average of two superimposed measurements.

At the core this material has a small ϕ-angle of 2° and the optical orientation of monoclinic orthoclase. Approximately halfway to the core the ϕ-angle is 5°.5, and the optic plane has undergone a clockwise rotation and approaches a position parallel to (010). At the edge the ϕ-angle is 6°, and the optic plane has rotated somewhat past a position parallel to (010). The angle $2V$ is small, approximately 24° from core to edge. Thus this

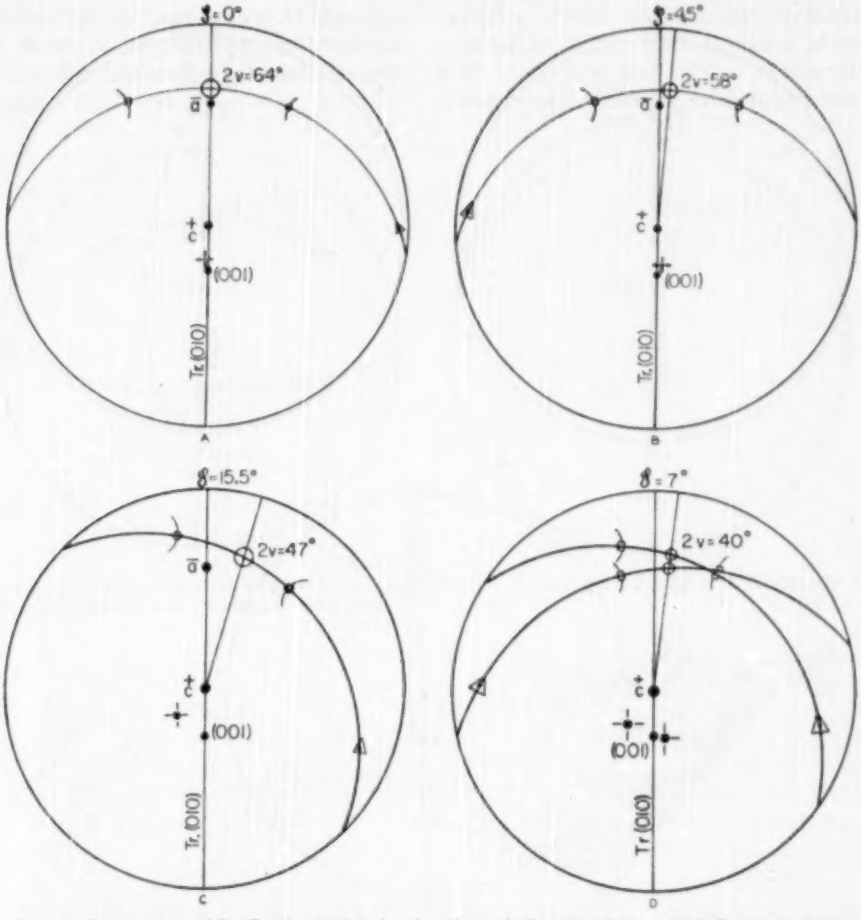

FIG. 5.—Stereograms of St. Gotthard adularia. *A*, a tiny, wholly monoclinic crystal; *B*, optical orientation at the core of a larger crystal; *C*, orientation at the edge of the same crystal as *B*; *D*, orientation of optic planes in a pair of albite twins, showing clockwise and anticlockwise rotation of optic planes around a.

material shows a complicated combination of optical properties. The core has the orientation of orthoclase, and the $2V$ of sanidine; the edges show monoclinic extinction angles and an orientation and $2V$ approaching, but not duplicating, those of sanidine.

Figure 7 illustrates the optics of the adularia known as "valencianite" from the Valencia silver mine at Guanajuato, Mexico. This material is twinned, as is that from Mount Vesuvius, after the albite law, with (100) the composition plane. Here again the (100) plane is somewhat irregular in the zone of the c-axis. The sketch of the crystal (fig. 9) emphasizes the peculiar blotchy appearance which may be caused by poorly developed pericline twinning.

Figure 7, *A*, illustrates the albite twin relationships of areas *1* and *2* of figure 9. Each of these two individuals appears

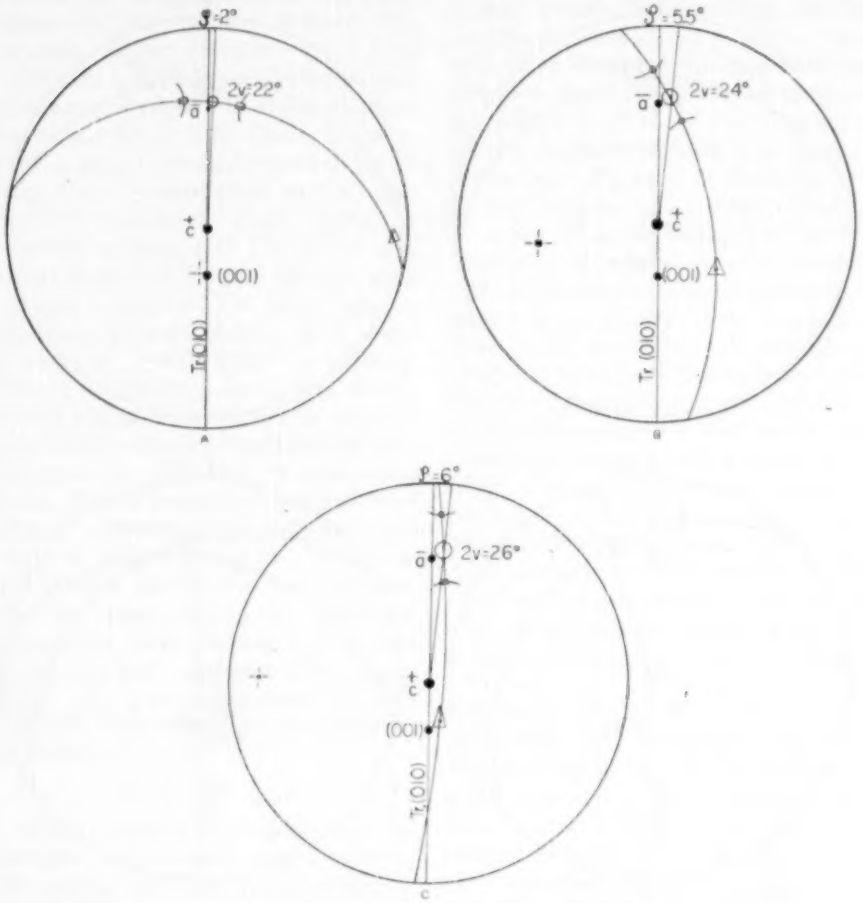

FIG. 6.—Stereograms of adularia from Mount Vesuvius. *A*, area *1* in fig. 6; *B*, areas *2* and *3*; and *C*, areas *4* and *5*.

essentially monoclinic. The angle ϕ is $0°$–$\frac{1}{2}°$, the optic plane is practically parallel to (010), and $2V$ is $45°$ (−). Figure 7, *B*, represents the average optical behavior of ten areas measured upon and near the edges of the crystal. All these areas were optically very similar. The average ϕ is $6°$, the average $2V$ is $56°$, and the optic plane is nearly parallel to (010). Thus, at the core, the valencianite specimen has albite twins with essentially monoclinic sanidine optics. At the edges it shows albite twinning, with triclinic extinction angles and the sanidine optic angle.

Measurements have been made of numerous adularia samples from these and other localities. In a general way the patterns outlined above are followed by all the other samples studied. For purposes of ready comparison, table 1 lists ϕ-angles, $2V$, and the orientation of several specimens.

CONCLUSIONS

The optical behavior of adularia is complex. Some crystals are optically monoclinic, some triclinic, some show both monoclinic and triclinic optics. Of those crystals showing both monoclinic

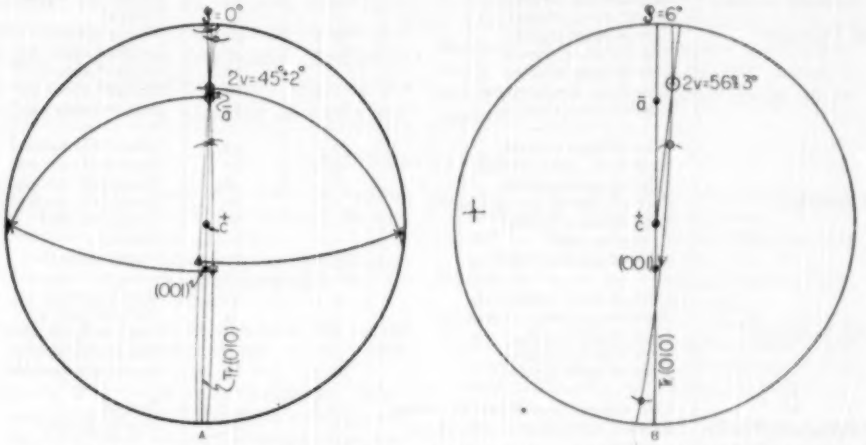

FIG. 7.—Stereograms of adularia from Guanajuato, Mexico. *A*, areas 1 and 2 of fig. 9; *B*, the average of ten areas measured near the edges of the crystal.

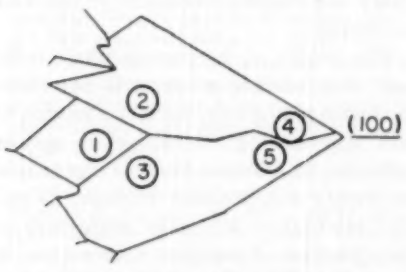

FIG. 8.—Sketch of adularia crystal from Mount Vesuvius, showing areas measured and described in text.

FIG. 9.—Sketch of adularia from Guanajuato, Mexico.

and triclinic optical behavior, some show a gradual, progressive change of properties. Others change along sharp boundary lines. The present study shows that, in general, the smallest (approximately 1–2 mm.) crystals of adularia show wholly or nearly monoclinic optics. Larger crystals (3 mm. and larger) commonly show extinction angles which adularia resembles microcline in the size of its axial angle more closely than does any other member of the orthoclase family. But Spencer (who makes no mention of triclinic adularia) does not suggest that there is any connection between the two. On the other hand, observations of the extinction angles in triclinic adularia convinced Mallard that all ortho-

TABLE 1

Locality	Part of Crystal Measured	ϕ (In Degrees)	$2V$ (In Degrees)	Orientation of Optic Plane to (010)
St. Gotthard	Single, small crystal	0	64	Normal
	Single, small crystal	2	30	Essentially normal
	Single, small crystal	4	30	Essentially normal
	Single, small crystal	0	.....	Normal
	Core of large crystal	4.5	58	Nearly normal
	Edge of same crystal	15.5	47	Inclined about 45°
	Core of large crystal	1	22	Inclined about 15°
	Near core, same crystal	1.5	22	Inclined about 15°
	Edge of same crystal	12	36	Inclined about 45°
Tavetschtal	Core of large crystal	0	30	Essentially normal
	Near core, same crystal	3.5	62	Essentially normal
	Edge of same crystal	5	36	Essentially normal
	Core of crystal (av. of 4 areas)	2.5	50	Essentially normal
	Edge of same crystal	5.5	28	Nearly parallel
	Core of crystal	1	.....	Normal
	Edge of same crystal	5	.....	Nearly normal
Mt. Vesuvius	Core of large crystal	2	22	Nearly normal
	Near core of same crystal	5.5	24	Inclined past 45°
	Edge of same crystal	6	25	Essentially parallel
	Core of large crystal	7	35	Essentially normal
	Edge of same crystal	11.5	35	Essentially parallel
Guanajuato Mexico	Core of large crystal (av. of 2 areas)	0–½	45 ± 2	Parallel
	Edge of same crystal (av. of 10 areas)	6	56 ± 3	Nearly parallel

are small toward the core but increase toward the edges of the crystal. The largest extinction angle seen by the writer in triclinic adularia crystals oriented normal to [001] approximates the extinction angles of microcline. It is evident that extremely small structural differences, some of which are too small to be measured by X-ray methods, cause striking changes in the shape and orientation of the indicatrix within the same crystal. According to the classification of Spencer (1937, his fig. 1), monoclinic clases are submicroscopically twinned microcline.

The results of the present study indicate that triclinic adularia is not identical with microcline, nor is it a modification approaching microcline in optical behavior. As adularia loses its monoclinic symmetry and becomes triclinic, its optical orientation and axial angle vary in the direction of sanidine rather than of microcline. This change toward the optics of sanidine, however, does not always follow the same pattern as that of mono-

clinic adularia and ordinary orthoclase when sanidinized by heat treatment. Heat treatment induces a change in orientation and optic angle, without the formation of extinction angles such as are seen in triclinic adularia.

It seems probable that adularia crystallizes in the monoclinic form and that a change to triclinic symmetry takes place later. This change begins at the acute angles of the crystal and progresses from the edges toward the core. During this change the optic plane of some crystals departs from its original orientation perpendicular to (010) and essentially parallel to (001) to become more and more steeply inclined to both planes and finally to become essentially parallel to (010) and normal to (001). This is accomplished by a rotation of the optic plane about the *a*-axis of the indicatrix and also approximately about the crystallographic *a*-axis, which is at all times very close to **a**. This rotation, when only partly accomplished, produces the triclinic extinction angles observed in adularia as it changes optically toward sanidine.

Triclinic adularia occurs as a secondary modification of monoclinic adularia. It has distinctive optical properties and behavior which establish it as a variety of potash feldspar.

ACKNOWLEDGMENTS.—This work was done in conjunction with a research program financed by the Geophysics Branch of the Office of Naval Research. The optical study was made concurrently with X-ray studies by Fritz Laves, to whom the writer is indebted for advice and cooperation.

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THE LATTICE AND TWINNING OF MICROCLINE AND OTHER POTASH FELDSPARS[1]

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ABSTRACT

The literature on potash feldspar (including the modern textbooks) gives one the impression that, within the limits of observation, the several varieties of $KAlSi_3O_8$ have the same apparent lattice "symmetry" (α and γ of microcline being quoted as 90° within the limits of $\pm 10'$). Two principal opinions exist to explain the different modifications: (1) The atomic arrangement (structure) of all potash feldspar is the same, and this structure is triclinic. The differences in optical behavior are explained by differences in the size and shape of twinned portions in the material (Mallard's hypothesis). (2) Two different modifications of potash feldspar exist: triclinic microcline and monoclinic orthoclase. It has been suggested that this difference is due to a different degree of order-disorder between the Si and the Al atoms, triclinic microcline having a greater degree of order than monoclinic orthoclase (Barth's hypothesis).

In this investigation it is shown that Mallard's hypothesis can be excluded, although it was here found that, optically, "monoclinic" potash feldspar composed completely of finely twinned microcline does exist. However, a new X-ray determination of the lattice of *microcline* ($\alpha = 89\frac{1}{2}°$, $\gamma = 92\frac{1}{2}°$) and the determination of the twin laws (albite, pericline) and their somewhat complex relations to each other, here discovered in microcline, indicate that such material originally grew as a monoclinic modification.

In addition, it has been found that another triclinic modification of $KAlSi_3O_8$ exists ($\alpha = 90°$, $\gamma = 90\frac{1}{2}°$), which is here called "triclinic adularia."

The structural relations between microcline, triclinic adularia, and "monoclinic" orthoclase are discussed, making use for the most part of intensity differences in the X-ray photographs of these three substances. Interpretation of the X-ray photographs of all "monoclinic" orthoclase investigated indicates that the monoclinic modification of potash feldspar is unstable at low temperatures. These photographs show diffuse streaks that are explained by partial transformation to a triclinic modification, which may be either microcline, triclinic adularia, or both.

INTRODUCTION

Des Cloizeaux (1875) first described two optically distinguishable potash feldspars: (1) orthoclase, optically monoclinic, and (2) microcline, optically triclinic. Mallard (1876) suggested that these were not distinct modifications but merely different developments of the same modification. Orthoclase was considered as a submicroscopically twinned microcline. Mallard reached this conclusion after observing that the size of the twin lamellae varies remarkably from place to place in the same crystal and that the twinning disappears completely in some places, such areas appearing optically monoclinic. This phenomenon could be seen in the photomicrographs published by Des Cloizeaux.

Several attempts have been made since to evaluate Mallard's hypothesis. Two approaches using X-ray techniques follow.

1. Barth (1929) and Schiebold (1928) investigated the symmetry of the microcline lattice. Their measurements (to be corrected later in this paper) showed no deviations of the angles α and γ from monoclinic symmetry within their limits of error ($\pm 10'$). Barth's conclusion was that, in so far as X-ray evidence was concerned, the lattice of microcline was the same as that of "monoclinic" orthoclase and that there is no reason to assume two modifications of K-feldspar. He stated that all potash feldspar is triclinic in structure but pseudo-monoclinic in lattice and that the optical difference is due to different kinds and degrees of twinning. X-ray work, therefore, seemed to

[1] Manuscript received May 20, 1950.

prove that Mallard's hypothesis was correct and that only one modification exists.

2. Another possibility was discussed by Barth (1934) after Taylor published his first determination of the structure of orthoclase. Taylor (1933) found a "monoclinic" unit cell containing four Al and twelve Si atoms, these atoms being situated in positions corresponding to two eightfold point positions, assuming the structure to be truly monoclinic. Taylor suggested that the real structure might be triclinic, in which case all four Al atoms would be situated in one of the four fourfold positions. Barth pointed out the possibility that the four Al and the twelve Si atoms are distributed at random ("variate atom equipoints"), thus forming a truly monoclinic structure; and he showed that the orthoclase-microcline relations could be explained by the assumption that orthoclase is truly monoclinic with a disordered distribution of the Al and Si atoms, whereas microcline is an ordered structure, truly triclinic but pseudo-monoclinic in so far as the limit of error ($\pm 10'$) of experimental X-ray work is concerned. This, in contrast to hypothesis 1 above, would indicate that potash feldspar exhibits polymorphism.

No experimental work has been done on this point since 1929 (Barth), and the question of whether only one form (triclinic) or polymorphic modifications (triclinic and monoclinic) of $KAlSi_3O_8$ exist has, to date, remained uncertain.

Taylor's structural determination of potash feldspar, combined with the hypothesis of order-disorder suggested by Barth, makes numerous structural types theoretically possible. Table 1 lists the six main possibilities. (It should be noted, however, that all the four fourfold positions of triclinic symmetry may be varied with respect to the whole structure, a point not indicated in the compilation. Thus types 4, 5, and 6 may each be split into several subdivisions.)

Barth's (1934) suggestion, discussed as hypothesis 2 above, apparently indicates that microcline is constituted as no. 6 in table 1, nos. 1 and 2 being reserved for monoclinic orthoclase.

TABLE 1

WAYS IN WHICH THE 4 AL AND 12 SI ATOMS CAN BE DISTRIBUTED IN THE BASE-CENTERED UNIT CELL OF K-FELDSPAR*

	No. of Equivalent Points per Unit Cell That May Contain 12 Si and 4 Al Atoms			
	Monoclinic Case			
	8		8	
1	Si+Al		Si+Al	
2	Si		Si+Al	
	Triclinic Case			
	4	4	4	4
3	Si+Al	Si+Al	Si+Al	Si+Al
4	Si	Si+Al	Si+Al	Si+Al
5	Si	Si	Si+Al	Si+Al
6	Si	Si	Si	Al

* The cell contains four "molecules" of $KAlSi_3O_8$.

In view of the uncertainty on the question of polymorphism in potash feldspar, it seemed desirable that additional evidence be found. If one assumes Mallard's hypothesis to be true, the "monoclinic" crystal would consist of thin layers with alternating structures. As is known from work on other substances with similar arrangements of alternating layers, the X-ray effects characteristically differ from those observed with normal crystals (for example, see Wilson [1943], Jagodzinski and Laves [1947]). What one observes are spots on the X-

ray photographs that show diffusion of intensity in the direction perpendicular to the layers. In the special case of exceedingly fine albite twinning, one would expect diffusion of spots in the direction of the b^*-axis.[1] The location and intensity of those spots which show diffusion will depend on the structure of the layers and their relations to one another. The size and the intensity of the diffuse spots depend on the thickness of the layers and on the amount of structural difference as produced by the difference in position in the alternating layers.

Although the structure of microcline has not been determined, it appeared to the writer that a study of comparative X-ray phenomena in microcline and "monoclinic" orthoclase might give at least some qualitative information. This investigation is divided into the following three sections: (*a*) the lattice and twinning of microcline, (*b*) the lattice of triclinic adularia, and (*c*) diffuse X-ray phenomena produced by "monoclinic" adularia. Conclusions that can be drawn from experiments described in these sections are here summarized.

1. The lattice of microcline deviates remarkably from monoclinic symmetry (γ is approximately $92\frac{1}{2}°$).

2. The well-known cross-grating twinning of microcline can be explained only by the assumption that such microcline originally crystallized as a modification with true monoclinic symmetry. It therefore appears that potash feldspar forms a true monoclinic modification under certain conditions. Whether or not a stability field exists for the monoclinic form is not yet known.

3. The "monoclinic" portions of those crystals that also show typical microcline twinning are ideal examples of Mallard's hypothesis. They are indeed submicroscopically twinned microcline. However,

[1] An asterisk after any symbol in this paper refers to the corresponding direction or plane in reciprocal space. Thus b^* = reciprocal b-axis = the normal to (010) of the original lattice.

PLATE 1

Precession photographs of twinned and untwinned (*E*) microcline. The precession axis is the *c*-axis. Cu-radiation was used in pictures *A–D*, Mo for *E* and *F*. The direction of a^* and b^* is shown in *A*. The (*hk*o) indices can be taken from fig. 5.

A, Microcline with albite twinning, Hunoholmen, Norway. Note that b^* is the same for both twins but that a^* is not. The angle between the two a^* positions represents the geometric deviation from monoclinic symmetry.

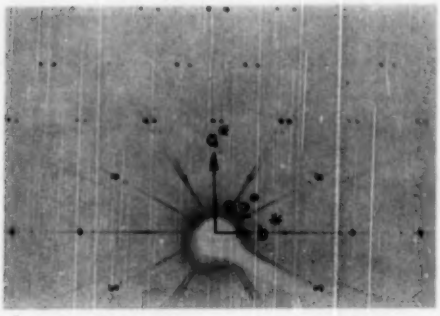
B, Microcline, Hunoholmen, Norway, portion that appears optically monoclinic. The photograph shows albite and pericline twinning, with all gradations between albite twin positions.

C, Microcline, Madagascar. This sample was optically homogeneous, with inclined extinction. No twin lamellae could be seen under the microscope. The photograph shows (compare with *A*) that albite twinning is actually present, one set of lamellae being much more abundant than the other (indicated by arrows).

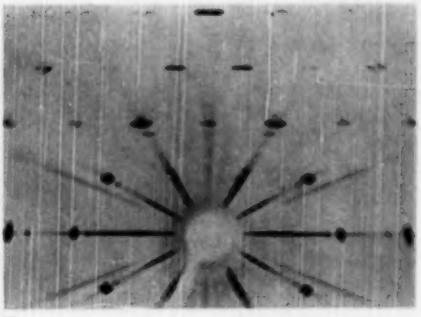
D, Microcline, Madagascar, portion that appears optically monoclinic. Definite spots in the monoclinic position are present, as are fainter albite twin spots. Diffuse streaks connect the albite position and the monoclinic spots.

E, Microcline, Hunoholmen, Norway, single crystal. Observe intensity differences between (*hkl*) and (*hkl*). These differences are greatest for those spots within the triangle and marked by the arrows. Figure 5 contains spot indices. (480), (390), (370), (190), have greater intensity than (480), (390), (370) and (190). The reverse is true for (170), indicated by the arrow.

F, Microcline, Hybla, Ontario, with cross-grating twinning (albite and pericline). The spots correspond to the arrangement in fig. 1, *C*. Notice the diffuse lines between (A_1, P_1) albite and pericline spots, geometrically described in fig. 2. The same intensity differences as in *E* are seen. Compare the spots in the triangle and the indices schematically shown in fig. 3. The streak indicated by the arrow should be compared with the corresponding streak in pl. 2, *E*, and is explained in the legend of fig. 1, *B*.



A



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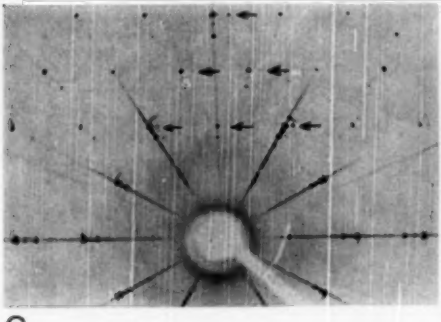
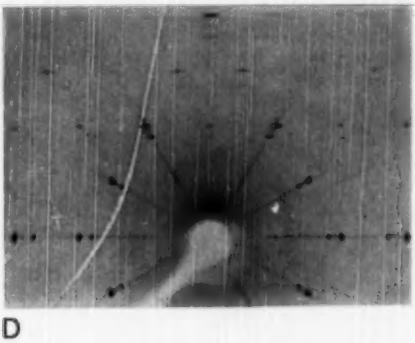
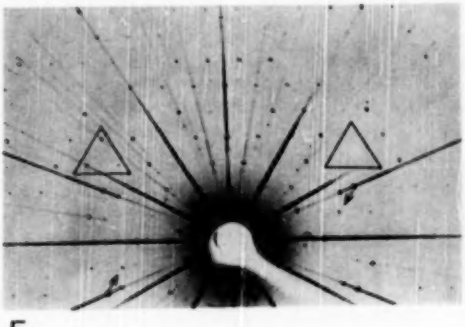


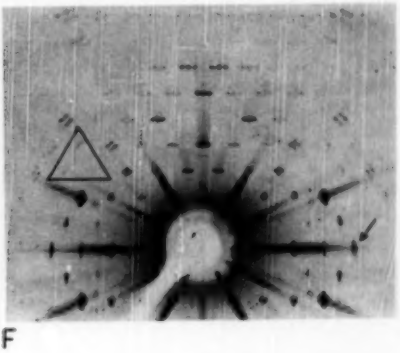
C



D

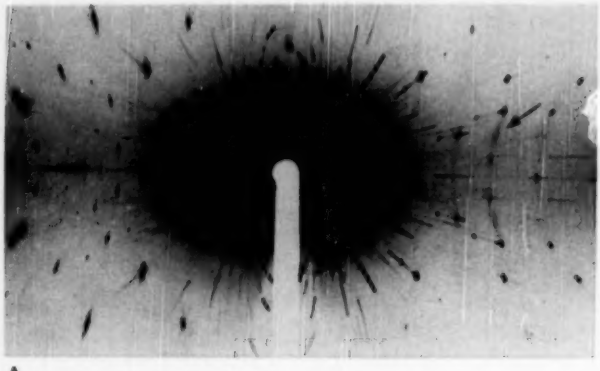


E

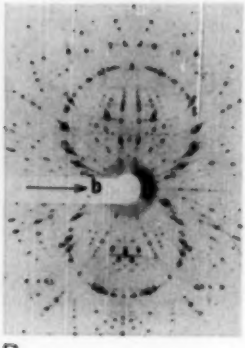


F

X-ray precession photographs of microcline



A



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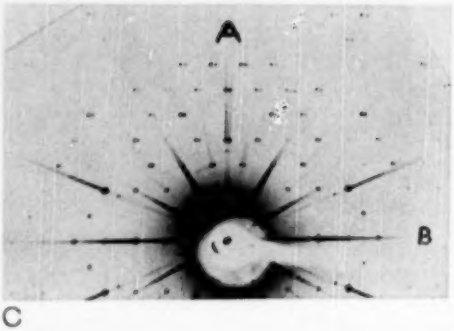

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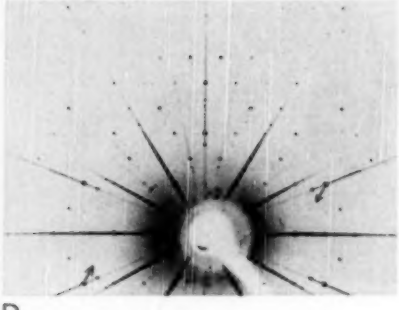
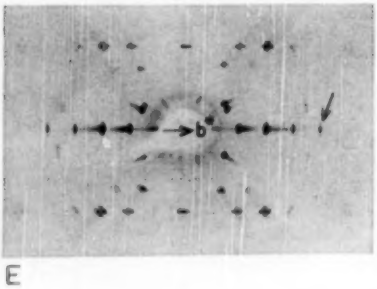


D



E

X-ray photographs of adularia ("monoclinic" and "triclinic")

other optically "monoclinic" material, for example, a very clear adularia from St. Gotthard, gave somewhat different results. In X-ray photographs no definite spots, sharp or diffuse, are present in locations characteristic of the microcline lattice. There are sharp spots, however, of intensity and position that correspond to monoclinic symmetry. In addition, there are diffuse streaks that indicate

TABLE 2

DATA IN THE LITERATURE ON THE GEOMETRY OF MICROCLINE*

No.	α	γ	(010)∧(001)	(010)∧(100)	Locality	Author
1	.....	.....	89°44′	.....	Pikes Peak	Des Cloizeaux (1875)
2	90°7′	89°55′	89°53′	.....	Hirschberg	Klockmann (1884)
3	.....	.....	89°32′	.....	Gasern	Sauer and Ussing (1890)
4	89°21′	91°59′	89°46′	88°7′	Pikes Peak	Böggild (1911)
5	89°19′	92°10′	89°43′	87°56′	Ivigtut	Böggild (1911)
6	90°7′	89°55′	.....	.....	Different localities	Schiebold (1928)
7	90°±10′	90°±10′	90°±10′	90°±10′	.....	Barth (1929, 1934)

* β is approximately 116°.

TABLE 3

NEW DATA ON THE GEOMETRY OF MICROCLINE AS DETERMINED IN THIS INVESTIGATION*

No.	(010)∧(100)	(010)∧(001)	γ	α	Locality
1	87°40′	89°35′	92°22′	89°18′	Hunoholmen
2	88°00′	89°38′	92° 3′	89°26′	Madagascar
3	87°50′	89°40′	92°15′	89°19′	Skede
4	87°47′	.....	.....	.....	Hybla

* β is approximately 116°. The maximum limit of error is ± 5′ for nos. 1 and 4, and ± 8′ for nos. 2 and 3.

PLATE 2

A, Oscillation photograph of "monoclinic" adularia (8° oscillation range around the *b*-axis, Fe-radiation). The spots on the right side are sharp, and between them are broad, diffuse spots, indicated by an arrow. On the left side the spots are accompanied by diffuse streaks, these streaks being fairly sharp in the directions normal to b^* (normal to streak elongation).

B, Laue photograph of "monoclinic" adularia, perpendicular to (100). Notice the diffuse lines parallel to $b^* = b$.

C, Precession photograph of triclinic adularia (sample no. 1 in table 6). Mo-radiation. The orientation is the same as in the photographs of pl. 1. Comparison of this photograph with pl. 1, *A* (microcline, albite twinning), shows the considerably smaller deviation from monoclinic symmetry in the case of triclinic adularia.

D, Precession photograph of triclinic adularia (sample no. 3, table 6). Mo-radiation. This photograph shows differences in (hkl) and $(h\bar{k}l)$ intensities in the same direction as in microcline, but comparison with pl. 1, *E*, shows that the effect is quantitatively smaller in triclinic adularia. This is well seen in the (170) and (1$\bar{7}$0) reflections, indicated in both photographs by arrows.

E, Precession photograph of microcline with cross-grating (same sample as in pl. 1, *F*). Here [104] was used as the precession axis. Mo-radiation. See fig. 5, *B*, for indices. (The additional spots not included in the schematic drawing of fig. 5, *B*, belong to other levels of the reciprocal lattice and may be neglected here.) Compare the streak marked by an arrow with the corresponding streak in pl. 1, *F*, and note that it is perpendicular to b^* when [104] is (as here) used as the precession axis. This phenomenon is explained in the legend of fig. 1, *B*.

crystal imperfections explainable as triclinic areas submicroscopically twinned. These results are suggestive of original growth in a monoclinic modification, with a subsequent transformation of small areas to a form with triclinic symmetry.

4. These areas appear to be related to the atomic arrangement of another modification of potash feldspar, one that may be called "triclinic adularia." Examination of other adularia crystals showed definite triclinic symmetry, with a γ of approximately $90\frac{1}{2}°$. The intensity differences of (*hkl*) and (*hkl*) in triclinic adularia are qualitatively (but not quantitatively) the same as in microcline. This fact might indicate that its atomic arrangement is one of groups 3, 4, and 5 shown in table 1, forms not so well ordered as that assumed for microcline.

The Lattice and Twinning of Microcline

The lattice of microcline

The values in table 2 for the crystallographic constants of microcline appear in the literature. Values 1–5 were obtained by optical and goniometrical methods, making use of the angles between cleavage planes (001), (010); the faces (010), (110), ($\bar{1}$10); and the "rhombic section." Values 6 and 7 were obtained by X-ray work, with an estimated accuracy of $\pm 10'$; and on the basis of these measurements it was claimed that no evidence exists for a difference between a "monoclinic" lattice and the lattice of microcline (Barth, 1928, 1929). Modern textbooks (Winchell, Strunz) have accepted these X-ray data and rejected the older goniometric measurements of Böggild.

A redetermination of these values with X-rays has been made by the writer, also with a maximum error of $\pm 10'$. No microcline was observed with γ less than 92°, as shown very clearly in plate 1. Table 3 lists the results of the present investigation.

As will be seen later, the exact measurement of the angles α and γ are important to the discussion of the twin laws in microcline and their relation to the question of the existence of a truly monoclinic modification of potash feldspar. In the following four sections some details of the substances investigated are presented.

Description of materials used

Microcline, Hunoholmen, Norway.—A crystal presumed to be albite (from a fissure, crystal $\sim$ 2 cm. long) proved to be composed of albite and potash feldspar when examined under the microscope. The potash feldspar shows optical variation of three principal types, with, however, all intermediate states: (*a*) optically "monoclinic" material; (*b*) optically triclinic, very finely twinned material (extinction between the twin positions not sharply defined); and (*c*) large areas, quite clear and with sharp asymmetrical extinction. The indicatrix is characteristic of microcline.

Plate 1, *E*, is a precession[3] picture of the (*hk*o) plane of the reciprocal lattice, the crystal being a portion of the *c*-zone (above).[4] It is immediately obvious that the angle (010)$\wedge$(100) = γ^* deviates appreciably from 90°. The exact value is given in table 3.[5] If a twinned

[3] See Buerger (1944) for a description of the precession technique.

[4] All the material investigated was thin-sectioned. After optical examination, a tiny portion of the section was carefully removed and mounted on the X-ray goniometer head. The relationship between optical and X-ray orientation was carefully recorded.

[5] The accuracy of the method used depends upon the precision of the moving film camera, the quality of the crystal, the accuracy with which the crystal is oriented, etc. One can normally, without difficulty, attain a limit of error of $\pm 10'$. If greater precision is

crystal is used, better than normal measurements can be made. Plate 1, *A*, illustrates this with a photograph of a portion of area *c* in the crystal, this portion showing the albite twin law.

Plate 1, *B*, is a precession picture of a "monoclinic" portion (*a*) of the crystal. Plate 1, *B*, differs from *A* in the following ways: (1) the sharp spots of *A* appear as diffuse spots in *B;* (2) the spots are linked by diffuse lines in the direction of b^*; and (3) the spots have diffuse tails that lie on the circumference of circles with the same Bragg angle. These phenomena will be discussed later in the section on twinning.

Microcline, Madagascar.—This green material had the same qualitative X-ray and optical behavior as the Hunoholmen microcline. Portions were (*a*) optically monoclinic; (*b*) finely twinned, optically triclinic; and (*c*) optically triclinic without observable twin lamellae, but with an extinction angle that varied from place to place ("undulatory" extinction).

Without the use of X-rays the optical behavior of type *c* could be interpreted in two different ways: (1) The undulatory extinction is due to variations of deviations, within the material, from monoclinic symmetry. Thus the optical deviation from monoclinic symmetry is a measure of the structural deviation, the structure varying somewhat from place to place in the crystal. (2) The material can be considered to be submicroscopically twinned, but all portions have the same (triclinic) structure. A difference in extinction angle is produced by variation in the proportions of right and left twin positions. This corresponds to Mallard's hypothesis. Plate 1, *C*, indicates that in this crystal the second view is correct. The reflections display the same deviations from monoclinic symmetry as those shown in the Hunoholmen microcline (pl. 1, *A*). The ratio of the intensities of the twin spots indicates the relative quantities of right and left twin material.

An X-ray photograph of a "monoclinic" portion (*a*) is shown in plate 1, *D*. The reflections (*hkl*) and (*hkl*) that correspond to microcline with albite twinning are visible only as very weak intensifications of the ends of streaks that run in the direction of b^*. In the center of the streaks very strong spots are observed. These spots belong to a monoclinic lattice, and one can also observe (unfortunately, not so well on the prints as on the original X-ray films) that these spots show tails that go off in the direction of positions that would be characteristic of pericline twin spots. This will also be discussed in the section on twinning.

Microcline, Skede, Norway.—The sample used was removed from a thin section of a large (~ 1 cm.), rather homogeneous crystal that showed uniform inclined extinction, the extinction angle being characteristic of microcline. Only a few patches of cross-grating twinning and some perthitic exsolution were present. The X-ray photographs are practically identical with those of the microcline from Hunoholmen (*c*, pl. 1, *E*), although the spots are not so sharply defined. The angular values are given in table 3.

Microcline, Hybla, Ontario.—A precession photograph of this finely twinned material is shown in plate 1, *F*. All the spots can be explained by the interpenetration of four lattices of the microcline type, the single crystal picture of which is shown in plate 1, *E*. The interpenetration of these lattices is such that two of

necessary, it is advisable to take precession pictures in the (+) and (−) directions. Observation of both pictures then cancels out any angular instrumental errors. A limit of error of ±5′ is then easily obtained.

them are related by the albite law, the other two by the pericline law. The schematic drawing (fig. 1) explains these relations. The angular values are given in table 3.

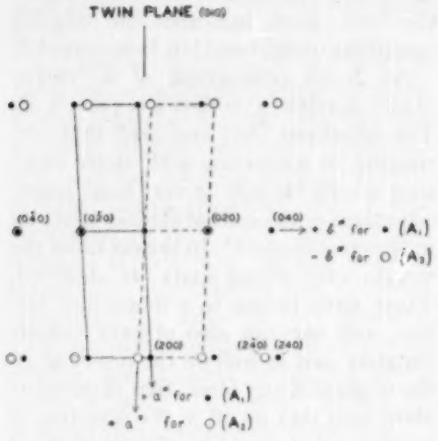


FIG. 1.—*A*, the (*hk*0) plane of two reciprocal lattices, A_1 and A_2, related by the albite law. The plane (010) is common to both twin positions, as is b^*, although the sign of b^* differs (+ and −) for the two positions. The angle between a^* and b^* corresponds to the angle between (100) and (010). To simplify this and the following diagrams, only the points for even values of h and k are included; the odd values of h and k are omitted. (The lattice of feldspar is base-centered, and thus only planes with even values of $[h + k]$ can be observed.) Because of this simplifying omission, one will observe additional spots on the corresponding photographs of pls. 1 and 2.

REMARKS ON THE TWIN LAWS IN MICROCLINE

All microclines investigated have the following features in common:

1. Deviations from monoclinic symmetry are, within a small limit, the same (see table 3):

$$(010)\bigwedge(001) = 89.^\circ6;\ \alpha = 89.^\circ4,$$
$$(010)\bigwedge(100) = 87.^\circ7;\ \gamma = 92.^\circ2.$$

2. As earlier investigators have assumed, the cross-grating twinning follows the albite and pericline twin laws,

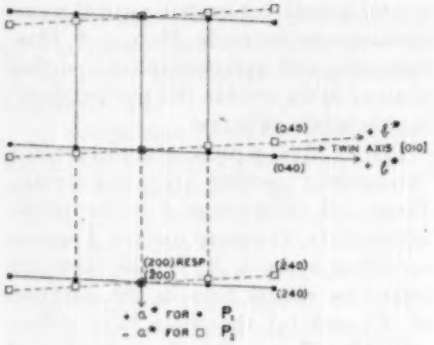


FIG. 1.—*B*, the (*hk*0) plane of two reciprocal lattices, P_1 and P_2, related by the pericline law. The twin axis $[010] = b$ (not b^*) is the same for both twin positions and, because (100) for P_1 corresponds to (100) for P_2, a^* is the same for both positions, although the sign of a^* differs (+ and −) for the two positions. The angle between a^* and b^* corresponds to the angle between (100) and (010). Again the points for odd values of h and k are omitted; only the even values of h and k are shown on the figure.

FIG. 1, *B* and *C* are only approximations of the true situation. The reciprocal lattice points of P_1 and P_2 will generally not fall in the same plane as drawn in *B*, because b^* for P_1, b^* for P_2, as well as a^*, are not generally situated in the same plane in the triclinic case. The following considerations show that these points do not lie in a plane. If $b^*(P_1)$, $b^*(P_2)$, and a^* did lie in a plane, it would follow that $[010]$ = twin axis = b would also lie in this plane. Because a^* and b^* are perpendicular to c (of the real lattice), b, lying in the plane of a^* and b^*, would also be perpendicular to c. If such were the case, α would be 90°. In triclinic material this would be a chance occurrence. Indeed, in the special case of microcline, $\alpha = 89.^\circ4$ (table 3). The precession technique is very sensitive to small disorientations, and this effect is quite obvious in precession photographs in which c is used as the precession axis. To digress briefly to fig. 1, *C*, we see that, if *A* (albite law) and *B* (pericline law) are combined so that b^* (of the albite figure) coincides with $[010] = b$ (of the pericline figure), a characteristic pattern is formed. In this pattern the pericline spots are drawn so that a line joining them is perpendicular to b (and b^*). Comparison of fig. 1, *C* with pl. 1, *F*, shows the agreement of the spot positions, neglecting the fact that the streaks crossing the b-direction (arrow in *F*) are not quite perpendicular to b. This angular difference is due to the fact that the angle between $c = [001] = [uow]$ and $b = [010]$ is 89.°4 and not 90°. If one chooses a direction $[uow]$ that is 90° from [010], and if this direction is used as the precession axis, a precession photograph will result in which the afore-

FIG. 1*B*.— *Continued*

mentioned streaks are perpendicular to $b = [010]$. A calculation using the determined values of α and γ for microcline (table 3) showed that a direction very close to [104] has an angle of 90° with [010]. A precession picture with [104] as the precession axis should therefore produce streaks perpendicular to [010]. Plate 2, *E*, shows such a photograph, which is proof that [104] is nearly perpendicular to [010].

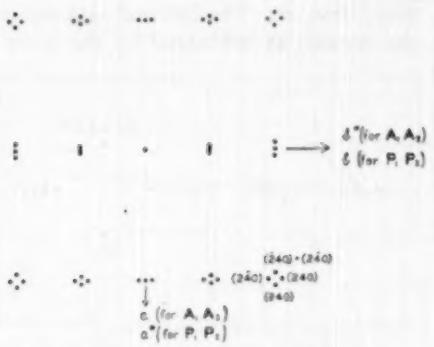


FIG. 1.—*C*, this figure illustrates the superposition of *A* and *B*. Here the spot positions in a plane (*hk*0) for a microcline twin composed of two albite positions, A_1 and A_2, and two pericline positions, P_1 and P_2, are oriented so that b^* for the albite twins coincides with b for the pericline twins. As in *A* and *B*, only the positions for even values of *h* and *k* are drawn. Close agreement exists between the pattern of *C* and the precession photograph of pl. 1, *F*. The correspondence of the twinning to the scheme of *C* is thus proved, taking into account the remarks under *B*.

but the real relations, unknown to date, were here determined.

3. In some materials, only albite twinning is observed. If pericline twins are present, albite twinning is also found; pericline twins have not been found alone.

4. If both kinds of twins are found, the following relations hold for the four positions A_1, A_2, P_1, and P_2 (fig. 1): A_1 and A_2 are related by the albite law, (010) is the twin plane; P_1 and P_2 are related by the pericline law, [010] is the twin axis. The twin axis is perpendicular to the twin plane. Although A_1 is related to A_2 by the albite law, and P_1 to P_2 by the pericline law, a rational relation between any *A* position and any *P* position does *not* exist because the material is triclinic. As explained in figure 2 and accompanying legend, the relation between any position *A* and any position *P* consists of an irrational rotation around an irrational direction.

It is difficult to understand how a growing crystal would follow such a complicated composition law. If one assumes that an albite twin A_1—A_2 forms (or one could start in reverse fashion with a pericline twin), the next step would be the formation of positions P_1 or P_2. It is exceedingly difficult to conceive of any forces that would cause position P_1 to assume a relationship to A_1 such that it deviates from A_1 by approximately a 3° rotation around an irrational direction near [104]. In a growing crystal, one would expect that, if a pericline position

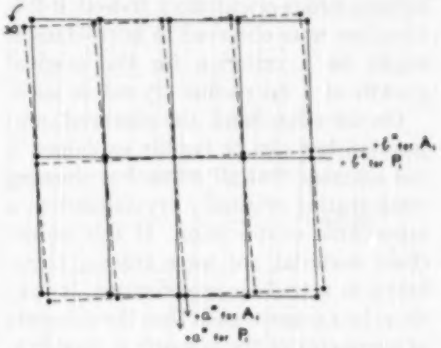


FIG. 2.—The relation between an A_1 (albite) and a P_1 (pericline) twin in reciprocal space fig. 1, *C*, illustrates the relations of the four twin positions, A_1 and A_2 (albite) and P_1 and P_2 (pericline), in reciprocal space, normal to the *c*-axis. Here an A_1 position from fig. 1, *A*, and a P_1 position from fig. 1, *B*, are combined, and it can be seen that a small rotation (approximately 3°) will produce coincidence of these two lattices. As pointed out under fig. 1, *B*, this rotation axis is not precisely the *c*-axis but an irrational direction close to [104]. The construction of this figure is therefore only an approximation for purposes of convenient illustration.

formed, it would follow the usual pericline law, showing a normal pericline relation to an already formed A_1 and A_2 albite twin. Thus, because a large deviation from monoclinic symmetry exists in microcline, a twin P_1 (related to A_1 by the pericline law) and a twin P_2 (related to A_2 by the pericline law) *would not show any twin relation to each other*. Carrying the argument further, if their parts P_1 and P_2 acted as partners to form additional albite twins, new positions A_1' and A_2' would be formed, *with no relation to* A_1 *and* A_2. This type of growth would form a patchy crystal, one with a highly heterogeneous orientation. Indeed, if this situation were observed in microcline, it might be a criterion for the original growth of a microcline crystal as such.

On the other hand, the observed composition law can be readily explained if one assumes that all microcline showing cross-grating originally crystallized as a monoclinic modification. If this monoclinic material, for some reason, transforms to a triclinic modification, it may do so in a manner such that the elements of symmetry of the monoclinic modification are preserved in twinned areas. Thus some areas would be twinned in albite positions A_1—A_2, retaining the plane of symmetry, and some areas would be twinned in pericline positions P_1—P_2, retaining the axis of symmetry. It is the writer's opinion that such a transition is required to explain the irrational composition law discussed above and shown in figure 2. No statement can be made here as to the stability of the monoclinic modification. It is not known whether it is stable at elevated temperatures or unstable under all conditions of pressure and temperature. An X-ray study of microcline, heat-treated for long periods of time, might clarify the stability relations; such a program is under way.

The photographs of plate 1 (*B* and *F*, in particular) illustrate differences in appearance that can be observed in X-ray photographs of microcline. These differences may depend on various factors governing the transformation from monoclinic to triclinic symmetry, such as temperature, pressure, composition (impurities), time, etc. The internal makeup of the crystal, as influenced by the inversion, is expressed in diffuse lines on the X-ray pictures. As shown in figure 1, *C*, the four reciprocal lattices A_1, A_2, P_1, and P_2 are interpenetrated, so that normally there are groups of four spots. The general relations between these spots are schematically represented in figure 3. In plate 1, *B*, *D*, and *F*, and plate 2, *E*, these diffuse lines or streaks can, for the most part, be seen joining the A_1—A_2 spots, on the one hand, and the A_2—P_2 and A_1—P_1 spots, on the other hand. These streaks can be qualitatively explained as follows: The monoclinic-triclinic transformation has not gone to completion, and sizable, well-formed triclinic portions are not de-

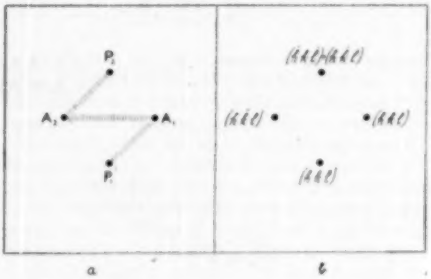


FIG. 3.—Appearance and indices of the spot groups in precession photographs of twinned microcline. Plates 1, *F*, and 2, *E*, show such arrangements. In figs. 1 and 2 the details of interpenetration of the four twin lattices were presented. Part *a* of this figure demonstrates more clearly than pls. 1, *F*, and 2, *E*, the position of the diffuse lines joining the spots A_1 and A_2 of the albite twin and P_1—A_1 and P_2—A_2, related by the approximately 3° rotation around the irrational direction near [104]. P_1 and P_2 are the pericline twin spots.

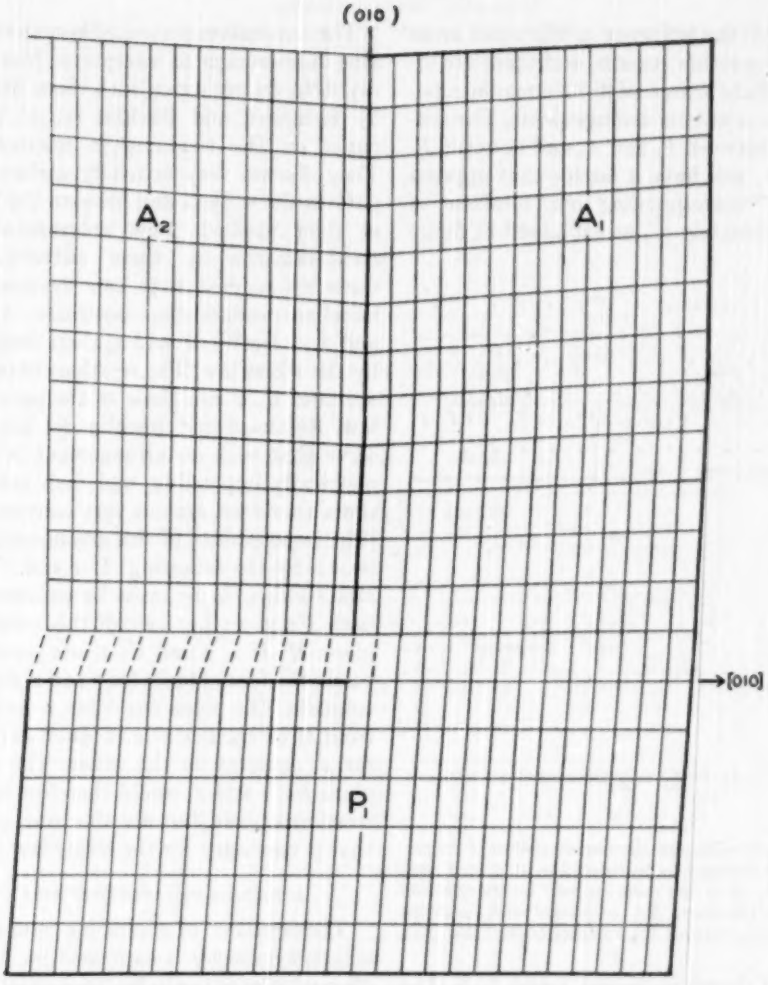


FIG. 4.—Model of the twinned lattices in microcline with cross grating twinning shown in a plane perpendicular to the *c*-axis. Again this model is an approximation for the same reasons as pointed out in figs. 1 and 2. The lack of rigor due to the two-dimensional presentation, however, is quite insignificant. This somewhat idealized model very well accounts for the X-ray phenomena here reported. A_1 and A_2 represent albite twin positions, and P_1 is one of two pericline positions. A_1 can easily change its orientation to that of P_1, but A_2 cannot. (Compare the distribution of the acute and obtuse angles of the parallelograms in the different positions.) This easy change from A_1 to P_1 is responsible for the well-developed diffuse lines seen connecting these spots on the X-ray photographs (see pls. 1, *B*, *F*, and 2, *E*).

The model also shows that dislocations exist between other twin parts ($A_2 - P_1$). Stress-strain relations must therefore exist in the twinned material. This stress, which is residual after most of the stress due to the crystal-transformation is eliminated by twinning, is kept at a minimum by very fine twinning. Most microcline is observed to be very finely twinned. This model is also consistent with optical observations on microcline. This deviation from monoclinic symmetry is exaggerated (4° instead of 2°) for the sake of clarity.

veloped; the influence of the small areas on one another remain, and there are intermediate states with intermediate positions or atomic arrangements. The material between P_1 and A_1 and between P_2 and A_2 will have a lattice that appears "bent," corresponding to a "rotation" of approximately 3°, as explained in figure

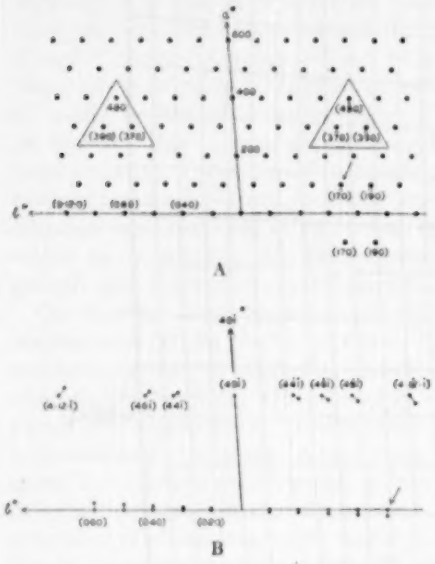


FIG. 5.—Diagram showing the indices of the precession photographs reproduced in pl. 1, *E*, *F*, and pl. 2, *E*. In *A* the spots inclosed by triangles and some of the others that are indexed show significant differences between the intensity of (*hkl*) and ($h\bar{k}l$).

2 and shown in figures 4 and 8, *B*. The material between A_1 and A_2 and between P_1 and P_2, on the other hand, will show less triclinicity than will normal microcline and, indeed, will pass through an orientation with true monoclinic symmetry (of at least *Cs-m* symmetry for A_1—A_2 and C_2-2 symmetry for P_1—P_2). Internal variation produces streaks that vary in definition and intensity from specimen to specimen, even though taken from the same crystal.

The conclusions presented here on twinning in microcline as interpreted from X-ray data do not agree with those drawn by Reinhard and Bächlin (1936) in a paper on the twinning in microcline. They discuss very critically earlier papers on the subject and bemoan the lack of rigor applied. New universal-stage measurements by these authors led them to suggest that two systems of lamellae produce four positions—1, 1′, and 2, 2′. Both 1, 1′ and 2, 2′ are twinned by the albite law. The relations between 1, 2 and 1′, 2′ are those of the pericline law. Reinhard and Bächlin did not observe that such an arrangement is geometrically impossible, and their conclusions, therefore, are not very convincing. [The impossibility of this arrangement is shown by the following: If 1 and 1′ are albite twins, (010) must be common to both 1 and 1′. Let us call this common plane *M*. If 1, 2 and 1′, 2′ are pericline twins, the (010) planes for 2 and 2′ do not coincide. The plane (010) for 2 deviates from *M* on the one side as much as (010) for 2′ deviates on the other. The twin plane for 2 and 2′ would therefore be an irrational plane, but not the (010) plane that is necessary for the albite law.]

A NOTE ON THE INTENSITY DATA

The deviation of microcline from monoclinic symmetry is expressed on X-ray photographs not only by the geometry of the lattice but also by intensity differences in some (*hkl*) and ($h\bar{k}l$) spots. Plate 1, *E* and *F*, shows these intensity differences. In table 4 some of the intensity data are presented. As a general rule it can be stated that (1) the average of the intensity of (*hkl*) and ($h\bar{k}l$) corresponds approximately to the (*hkl*) intensity of "monoclinic" potash feldspar and (2), when there are differences in (*hkl*) and ($h\bar{k}l$) intensities, the differences are larger

TABLE 4a

INTENSITIES OF (*hk*o) PLANES OF MICROCLINE, TRICLINIC ADULARIA, AND MONOCLINIC ADULARIA*

h =	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
(10*k*o)	5 5 5		oo oo oo		32 −1 22 22		33 22											
(9*k*o)		33 33 33		33 22 22		44 33 33		54 −1 44 44		55 55 55								
(8*k*o)	o o o		o3 +3 22 22		44 33 33		23 +1 22 22		45 +1 55 55		oo oo oo							
(7*k*o)		66 66 66		54 −1 55 55		oo 11 oo		33 22 33		33 33 33		13 +2 22 22		44 44 44				
(6*k*o)	7 6 7		44 55 55		44 44 44		45 +1 55 55		22 33 33		11 oo oo		oo oo oo		o3 +3 oo 11			
(5*k*o)		12 +1 1′1′ 22		55 66 66		44 44 44		11 1′1′ 22		55 55 55		10 −1 oo oo		oo oo oo		oo oo oo		
(4*k*o)	8 8 8		44 55 55		44′ +½ 44 44		55 55 55		o4 +4 23 +1 33		43 −1 44 44		oo oo oo		33′ +½ 33 33		44 44 44	
(3*k*o)		55′ +½ 66 66		33′ +½ 44 3′3′		55 55 55		o3 +3 o2 +2 o′o′		o4 +4 33 22		44 4′4′ 44		32 −1 22 22		oo oo oo		35 +2 4′4′ 55
(2*k*o)	6 5 6		88 88 88		55′ +½ 55 55		56 +1 44 55		88 88 88		10 −1 oo oo		23 +1 33 22		33 33 33		21 −1 oo oo	
(1*k*o)		41 −3 4′4 −½ 33		88 88 88		o2 +2 oo oo		40 −4 20 −2 oo		35 +2 45 +1 44		oo oo oo		oo 11 o′o′		oo′ +½ oo oo		12 +1 oo oo
(0*k*o)			5 4		8 8 8		8 8 8		6 5 6		6 5 6		o o o		1 1 1		5 5′ 5	

* Taken from precession photographs as in pl. 1, *E* (Mo-radiation).

NOTE TO TABLE 4a

The figures 0, 1, 2, 3, 4, 5, 6, 7, 8, represent intensity values of 0, 1, 2, 4, 8, 16, 32, 64, 128. The intensity values of the different modifications are arranged in the following way.

A	B
C	D
E	F

The *A* position is the value of (*hk*$\bar{o}$) in microcline; the *B* position is the value of (*hk*o) in microcline; the *C* position is the value of (*hk*$\bar{o}$) in tr. adularia; the *D* position is the value of (*hk*o) in tr. adularia; the *E* position is the value of (*hk*$\bar{o}$) in mcl. adularia; the *F* position is the value of (*hk*o) in mcl. adularia.

n′ signifies an intensity value between *n* and (*n* + 1). In some cases a difference in intensity between (*hk*o) and (*hk*$\bar{o}$) exists. Here these intensity differences appear as extra figures in the box. A (+) signifies (*hk*o) > (*hk*$\bar{o}$), and (−) signifies (*hk*o) < (*hk*$\bar{o}$).

for those spots of low intensity. The greatest intensity difference found on oscillation photographs (with Cu Kα-radiation) was of the order of 1:100, structurally the intensity differences. In the next section, however, some comparisons with another triclinic modification of potash feldspar will be made.

TABLE 4*b*

INTENSITIES OF (0*kl*) PLANES

h =	0	2	4	6	8	10	12	14	16
(0*k*7)	2 2 3	04 +2 22′ +½ 3′3′	4′2 −2½ 33 44	oo oo oo	01 11 1′1′				
(0*k*6)	o o o	3′1′ −2 33 33	oo oo oo	30 −3 2′2 −½ 2′2′	3′4′ +1 44 3′3′	oo oo oo	oo oo oo		
(0*k*5)	4 4 4	25 +3 34 +1 3′3′	62 −4 43′ −½ 4′4′	02 +2 22 1′1′	02 +2 22 1′1′	20 −2 oo oo	10 −1 oo oo		
(0*k*4)	.. 2 2	66′ +½ 66 66	oo oo oo	44 44 44	4′6 +1½ 55 55	1′o −1½ oo oo	11′ +½ 11 1′1′	oo oo oo	
(0*k*3)	4 2 3	oo oo oo	67 +1 55 55	44′ +½ 44 44	44 44 33	01 +1 oo oo	2′3 +½ 33 44	o′o′ oo oo	
(0*k*2)	8 8 8	65 −1 66 55	oo oo oo	66 55 55	4′o′ −4 43 −1 44	13′ +2 44 33	oo oo oo	13 +2 22 22	
(0*k*1)	4	40 −4 31′ −1½ o′o′	76 −1 66 55	45 +1 44 44	o′4 +3½ 33 33	10 −1 oo oo	43 −1 33 33	40 −4 10 −1 11	30 −3 10 −1 oo
(0*k*0)	.. .. ..	.. .. ..	8 7 6	8 8 7	5 4 4′	5 4 5	.. .. ..	.. .. ..	5 4 5

using *b* as the oscillation axis and including all planes with $l < 7$ and a Bragg angle $< 70°$. This figure is based on the examination of three hundred pairs (*hkl*) and (*hkl*) (table 5). These values appear to support Barth's (1934) suggestion that triclinic microcline differs in its degree of order from monoclinic orthoclase. For additional remarks on this subject see the companion paper by Goldsmith in this issue of the *Journal*. No attempt has yet been made to evaluate

Triclinic Adularia

In 1876 Mallard observed that some crystals of adularia from Switzerland deviated optically from monoclinic symmetry. Barth (1928) and Köhler (1948) observed an analogous phenomenon, but no X-ray studies were made. Mallard accepted this triclinic behavior as supporting evidence for his hypothesis that all monoclinic potash feldspar is submicroscopically twinned microcline.

A number of adularia specimens from

TABLE 5

INTENSITY RELATIONS OF ADULARIA AND MICROCLINE

A	B	C	D	E	F
Scale of Intensities Used	Actual Intensities of Figures in Col. A	No. of Planes of Adularia Measured with an Intensity Shown in Col. B	Each Plane of Adularia Summarized in Col. C Symbolized Here by a Number. Each Number Is a Measure of the Intensity Ratio of the (*hkl*)—($\bar{h}\bar{k}\bar{l}$) Planes in *Microcline* That Correspond to Equivalent (*hkl*) Spot in Adularia Summarized in Col. C.	Sum of Numbers in Col. D	Col. E Divided by Col. C
1	.....	3	4 4 1	9	.....
2	2	2	1 0	1	.....
3	.....	7	11 8 6 6 5 4 0	40	5.7
4	4	12	7 7 7 7 5 4 4 3 3 2 2 0	51	4.3
5	.....	11	8 7 7 4 4 3 2 2 2 1 0	40	3.6
6	8	6	6 5 5 4 4 0	24	4.0
7	.....	16	13 12 12 10 10 8 8 6 6 5 4 3 3 2 1 0	103	6.4
8	16	27	12 12 12 11 6 6 5 5 5 4 4 4 3 2 2 2 2 2 2 1 1 1 0 0 0 0 0	104	3.9
9	.....	19	15 13 8 5 4 3 3 3 3 2 1 1 1 1 1 0 0 0 0	64	3.4
10	32	25	12 12 6 5 4 4 4 4 4 4 4 3 3 3 3 3 3 3 2 2 1 1 1 0 0	91	3.6
11	.....	24	14 10 9 8 8 6 6 4 4 3 3 3 2 2 2 2 2 2 1 1 0 0 0 0	92	3.8
12	64	23	8 7 6 6 5 5 4 4 3 3 3 3 2 2 2 2 1 1 1 0 0 0 0 0	66	2.9
13	.....	18	8 5 5 5 4 4 2 2 2 2 2 1 1 0 0 0 0 0	43	2.4
14	128	27	8 7 4 4 4 4 4 3 3 3 2 2 2 2 2 2 1 1 1 0 0 0 0 0 0 0 0 0	59	2.2
15	.....	10	3 2 2 2 2 1 1 0 0 0	13	1.3
16	256	17	4 4 4 3 2 2 2 2 1 1 1 1 1 0 0 0 0	28	1.6
17	.....	10	4 2 2 2 2 1 1 1 0 0	15	1.5
18	512	9	2 2 2 2 2 1 0 0 0	11	1.2
19	.....	6	5 2 2 0 0 0	9	1.5
20	1,000	3	2 0 0	2	0.6
21	.....	2	0 0	0	.....
22	2,000	1	0	0	.....
Total	.....	278	.....	.....	.....

NOTE TO TABLE 5

The intensities of 278 (*hkl*) planes of "monoclinic" adularia were estimated from oscillation photographs made with *b* as the oscillation axis. Intensity estimations were made, using a previously exposed film standard. Three sets of oscillation photographs were taken, between which the exposure time was varied so that it was possible to distinguish intensity variations of from 1 to 2,000. Column A shows the standard scale of intensities and is related to col. B, which shows the actual intensity values. In col. C the number of planes measured (with the corresponding value in col. B) are tabulated. Column D illustrates the intensity differences that exist between (*hkl*) and ($\bar{h}\bar{k}\bar{l}$) in microcline (see column heading). For example, consider row 19, the spots in which have "actual" intensity values between 512 and 1,000. Column C shows that there were six spots measured with the intensity. To these six planes (*hkl*) of adularia, there correspond six pairs of (*hkl*)—($\bar{h}\bar{k}\bar{l}$) planes in col. D, with intensity ratios between the (+) and (−) spots of 5, 2, 2, 0, 0, 0. This signifies that one pair has an intensity ratio of about 6, two pairs a ratio of 2, and three pairs show the same (+) and (−) intensities. The largest difference of intensity between any (*hkl*) and ($\bar{h}\bar{k}\bar{l}$) spots observed in these oscillation photographs had the value 15 (see row 9), which corresponds to an actual intensity difference (see col. B) of about 200. This large intensity difference corresponds to a relatively small intensity in adularia. (Row 9 has an actual intensity of about 25.)

On the average, the larger (*hkl*)—($\bar{h}\bar{k}\bar{l}$) differences in microcline correspond to relatively small actual (*hkl*) intensity values in adularia, as illustrated by col. F. Additionally, it should be pointed out that the *average* (*hkl*)—($\bar{h}\bar{k}\bar{l}$) intensity of microcline for the most part is similar to the equivalent (*hkl*) for adularia.

each of several localities (Switzerland, Vesuvius, Mexico) were examined optically and with X-rays. The description and optical measurements are given in a companion paper by Chaisson in this issue of the *Journal*.[6] The X-ray work has proved that this material is distinct from microcline and must be regarded as a separate modification. All samples used in the X-ray investigation were taken from sections after they had been measured optically on the universal stage. In all cases the relation between the position of the optical indicatrix and the geometrical ones (as determined by X-rays) was recorded. This was not very easily done in all cases because of the lineage structure present in most of the crystals and because of their small deviations from monoclinic symmetry. The photographs were taken, for the most part, by the precession technique and with a limit of error as mentioned in the section on microcline. The lattice angles are given in table 6.

LATTICE ANGLES OF TRICLINIC ADULARIA

Comparison of these values for triclinic adularia with those of microcline (table 3) shows that the deviation from monoclinic symmetry is in the same direction as in microcline. The amount of deviation, however, is only about one-fourth that in microcline, as is evident from comparison of plate 2, *C* and *D*, with plate 1, *A* and *E*.

Another analogy is that qualitatively the intensity differences between (*hkl*) and (*hkl*) planes are the same in triclinic adularia as in microcline, although the effect is much less pronounced. Plates 2, *D*, and 1, *E*, illustrate the relative intensity differences. Table 4 indicates the

TABLE 6

DATA ON THE GEOMETRY OF TRICLINIC ADULARIA*

Sample No.	(010)∧(100)	(010)∧(001)	γ	α	$2V_a$ and Position of Opt. Axial Plane†	Locality
1	89°20′ ± 5′	89°50′ ± 5′	90°40′	89°52′	40°∼⊥	St. Gotthard
2	89°39′ ± 8′					Tavetschtal
3	89°26′ ± 5′	89°50′ ± 5′	90°33′	89°54′	26°∼	Vesuvius
4	89°28′ ± 5′	89°51′ ± 5′	90°32′	89°56′	26°∼	Vesuvius
	89°30′ ± 5′	89°54′ ± 5′	90°30′	89°52′	26°∼	Vesuvius after heating to 1,000° C.
5	89°38′ ± 7′				35°∼	Vesuvius
6	89°29′ ± 8′				56°∼	Mexico; "Valencianite"
7	89°33′ ± 5′	89°50′ ± 5′	90°24′	89°57′	52°∼	Mexico; "Valencianite"

* β is approximately 116°.

† The signs ∼⊥ and ∼‖ indicate the position of the plane of the optic axis relative to (010). As pointed out in the accompanying paper by U. Chaisson, the plane is neither parallel nor perpendicular to (010). Thus the signs merely indicate that the plane is located closer to one position than to the other.

Using the accepted system of orientation for the feldspars [the angle (010)∧(001) being smaller than 90°], the optical deviation from monoclinic symmetry in triclinic adularia is in the same direction as in microcline (a clockwise rotation around an axis close to the *a*-axis, when viewed in the positive direction).

Samples 3 and 4 represent measurements made on the same crystal as that described in the accompanying paper by U. Chaisson (see fig. 6, locations 4 and 5, in Chaisson's paper). The two measurements represent the two different twin positions. A sample taken from the core of this crystal (not listed in this table) was optically nearly monoclinic, and the X-ray pictures indicated monoclinic geometry within ± 5′. Monoclinic geometry was also observed in the core of a crystal which showed, near the edge, the triclinic values listed under no. 5.

A heat treatment of sample 4 for 100 hours at 1,000° C. produced no geometric change (within the limits of error).

[6] From the optical study, as discussed in the paper by Chaisson, it is apparent that a transformation from original monoclinic symmetry to the triclinic form is indicated in adularia.

order of magnitude of these intensity differences.

If one accepts the reasonable assumption that microcline is an ordered modification of potash feldspar with the atomic distribution of no. 6 in table 1, one is led to the conclusion that triclinic adularia represents one of the other three distributions, nos. 3, 4, and 5, in table 1.

Considering the fact that the deviation from monoclinic symmetry is small as compared to microcline and following the likely assumption that the transformation from monoclinic to triclinic symmetry probably takes place at rather low temperatures, it is the writer's opinion that the most likely atomic distribution of triclinic adularia is that of no. 3 in table 1. This would indicate a triclinic modification of $KAlSi_3O_8$ that is completely disordered.

There is further evidence that favors this hypothesis. In all investigated adularias from six different localities (four Swiss localities, Vesuvius, and Mexico) the triclinic modification was found. All show the same deviation from monoclinic symmetry ($\gamma = 90\frac{1}{2}°$) within a small limit. Microcline was not found as a part of, or associated with, any of the crystals. This fact might indicate that no interatomic diffusion or exchange is involved in the formation of triclinic adularia from the monoclinic modification. If diffusion were responsible, why should it stop after the formation of triclinic adularia? Certainly, in places it would be expected to have gone far enough to form a partly ordered material, if not microcline. Conversely, X-ray photographs show no evidence of the existence of triclinic adularia in microcline. A structural deformation requires less total atomic movement than does rearrangement to an ordered form by atomic exchange.

These suggestions imply that triclinic adularia is metastable. It should be completely ordered to microcline, but the formation temperature (and subsequent temperature at which the monoclinic-triclinic transformation took place) was too low to permit microcline formation.

"Monoclinic" Adularia

It was earlier shown that the ordinary cross-grating twinned microcline cannot be explained except by the assumption that the crystal was previously monoclinic. It was shown above that there are at least two different triclinic modifications of potash feldspar—microcline and triclinic adularia. It has been suggested that microcline is formed from monoclinic feldspar at temperatures high enough for atomic rearrangement by diffusion, whereas triclinic adularia is formed from monoclinic feldspar at relatively low temperatures. These assumptions would necessitate the formation of all adularia as a metastable monoclinic modification.

In the first section optically monoclinic orthoclases were described that showed fine triclinic X-ray twinning and thus corresponded to Mallard's conception of orthoclase. Such optically monoclinic material was, however, always observed to have adjacent areas with triclinic optics. On the other hand, those crystals of optically monoclinic feldspar in which microcline could not be detected gave X-ray pictures unlike the material that follows Mallard's hypothesis. The normally exposed pictures had sharp spots that corresponded to monoclinic symmetry, both in geometry and in intensity, the intensity of (hkl) being equal to that of $(h\bar{k}l)$. In long-exposed X-ray pictures some diffuse effects appeared that are explainable as imperfections in the monoclinic lattice due to triclinic areas preponderantly related by the al-

bite law. Some pericline areas are also present but are very minor in quantity and will not be further discussed. These areas are thin lamellae parallel to (010) and thus cause diffuse streaks parallel to b^* in the reciprocal lattice. Laue photographs show these effects with relatively short exposures (pl. 2, *B*). The material that gave the most pronounced diffuse patterns was an adularia from St. Gotthard.[7] This was used for further investigation. Oscillation photographs with *b* as the oscillation axis (Cu- and Fe-radiation) were made, to obtain more complete information on the diffuse phenomena in reciprocal space. Plate 2, *A*, is a picture that clearly shows two different diffuse phenomena: (1) one type is associated with the main spots and is fairly sharp in the directions perpendicular to b^*; (2) the other type has its most intense area between the spots and is rather diffuse perpendicular to b^* (see arrow, pl. 2, *A*). To date, the writer sees no clear explanation for the second type. These areas do, however, suggest regions in the crystal in which the base-centering of the normal feldspar structure is disturbed. Neither of these phenomena has been observed in either untwinned triclinic microcline or in plagioclase (which is also triclinic). They seem to be restricted to "monoclinic" orthoclase.

The first type of diffuse lines associated with the main spots follows characteristic rules, one of which is closely related to the geometry of the lattice of microcline and triclinic adularia. (1) The intensity of each diffuse area is a function of the intensity of its corresponding main spot. If the ratio of these two intensities be called *R*, then (2) *R* is a function of the spacing, *d*. The smaller the *d*-value, the larger is *R*. (3) *R* is also a function of the position of the corresponding main spot in the reciprocal lattice. Expressed in terms of the real lattice, *R* is close to zero for planes in the zone [401] and has a maximum value for planes nearly perpendicular to [401]. Planes with intermediate positions have intermediate *R*-values, dependent upon the angle between the plane and [401].

Figure 6 summarizes the results of intensity measurements on approximately six hundred spots.

These effects can be qualitatively explained by the statement that the principal part of the crystal behaves as a truly monoclinic structure as far as the geometry, intensity, and sharpness of the spots on the X-ray photographs are concerned. Within this monoclinic material are layers, preponderantly parallel to (010), with atomic arrangements that deviate somewhat from the principal structure. These layers produce the additional streaks, parallel to b^*, that are associated with the sharp monoclinic spots. Plate 3, *A*, *B*, shows these streaks quite clearly. Most of the streaks on the photographs hide the main spots, inasmuch as these pictures were deliberately overexposed in order to illustrate another important effect. Comparison of the left and right sides of *A* and *B* (pl. 3) also illustrates the fact that most of the spots on the left side are not accompanied by streaks, whereas those on the right side are. This signifies that the appearance of streaks is a function of the location of the

[7] This adularia, with chemical analysis, was kindly supplied by Dr. N. L. Bowen, and had the following composition:

	Per Cent		Per Cent
SiO_2	63.95	CaO	0.04
TiO_2	0.00	Na_2O	1.17
Al_2O_3	19.66	K_2O	14.43
Fe_2O_3	0.05	BaO	0.08
FeO	0.00	$H_2O(+)$	0.56
MgO	0.04	$H_2O(-)$	0.16

Or = 88.7, Ab = 10.9, An = 0.2

main spots in reciprocal space. The conclusion can thus be drawn that the layers producing the streaks are not without "coherent relations" to their environment and that there are directions in which the crystal behaves as a perfect one. In these directions the presence of the sheets does not disturb the X-ray reflections.

RELATION OF THE [401] ZONE IN ADULARIA TO THAT IN MICROCLINE AND TRICLINIC ADULARIA

Experimentally (see rule 3, above, and fig. 6) it has been found that the directions in which the crystal acts as a perfect one to X-rays are nearly perpendicular to [401]. It is striking that the [401] zone, unique with respect to the above

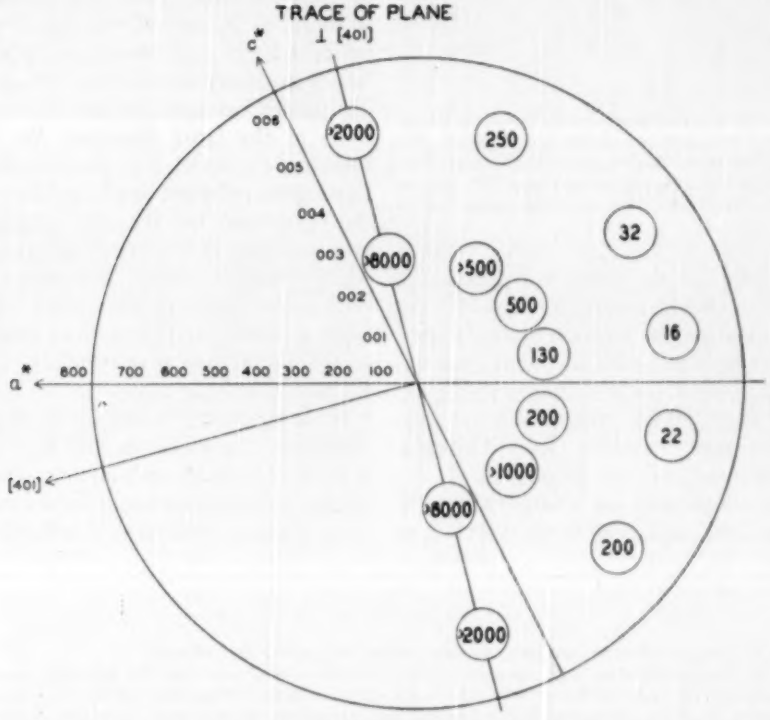


FIG. 6.—A projection of reciprocal space of adularia, on the plane perpendicular to b (in the monoclinic case $b = b^*$). This plane contains the reciprocal axes a^* and c^* and the points corresponding to the (h00) and (00l) spots. The (h0l) points are excluded for simplicity. The circled numbers represent intensity relations between the diffuse streaks and their associated main spots. They are average values taken from oscillation photographs (b = oscillation axis, oscillation range 8°, Fe-radiation), using (hkl) reflections, with l less than 6. The number in each circle represents the center of an area of reciprocal space containing approximately 20–30 spots.

Each number is specifically the average for that area of the ratio of the main spot intensities to those of the diffuse streaks. Thus small values indicate relatively high diffuse streak intensities and vice versa. The diagram shows that the intensities of the diffuse streaks are virtually zero for planes in the [401] zone and are greatest for planes nearly perpendicular to this zone. It can also be observed that the relative intensities of the diffuse streaks grow larger with greater distance from the zero point of the reciprocal lattice. Thus the smaller the spacings, the greater is the relative intensity of the diffuse streaks.

phenomenon, also has a unique significance in microcline and triclinic adularia, as the following discussion shows.

In any triclinic lattice there is one (generally irrational) plane (*hol*) that is perpendicular to (010). Its position is such that it must contain *b* and *b**. One can calculate the position of this plane; and, using α and γ of microcline and triclinic adularia (as determined in the investigation, tables 3 and 6), it is found to be very near the plane ($\bar{1}04$). The zone common to ($\bar{1}04$) and (010) is [401].

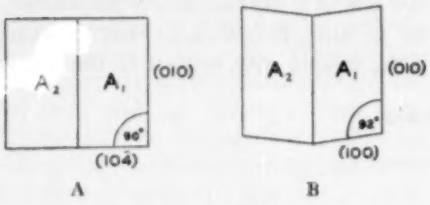


FIG. 7.—*A* is a schematic projection of the lattice outline of two albite twins in the [401] direction; *B* is a projection in the [001] direction. In the [401] direction the two twins appear as *one* crystal. The indices refer to planes which appear in the projections as lines.

Let us project an albite twin with planes ($\bar{1}04$) and (010) in the direction of their common zone [401] (fig. 7, *A*). Now let us compare this projection with one of an albite twin with the planes (100) and (010) in the direction of the *c*-axis (fig. 7, *B*). The difference is obvious. In the [401] direction we can see no difference in the two twin positions, but in the [001] direction the difference is apparent. This is a two-dimensional representation of deviation from monoclinic symmetry in figure 7, *B*, and of monoclinic symmetry in figure 7, *A*. Thus, in so far as the lattice geometry is concerned, we cannot distinguish between the two albite positions in the [401] direction. We may therefore expect little or no difference in those spots reflected by planes in or near the [401] zone. On the other hand, the two positions in the [001] direction are readily distinguishable, and we may expect some degree of diffuseness for all spots reflected by planes in or near the [001] zone (as long as these planes do not approach the [401] zone).

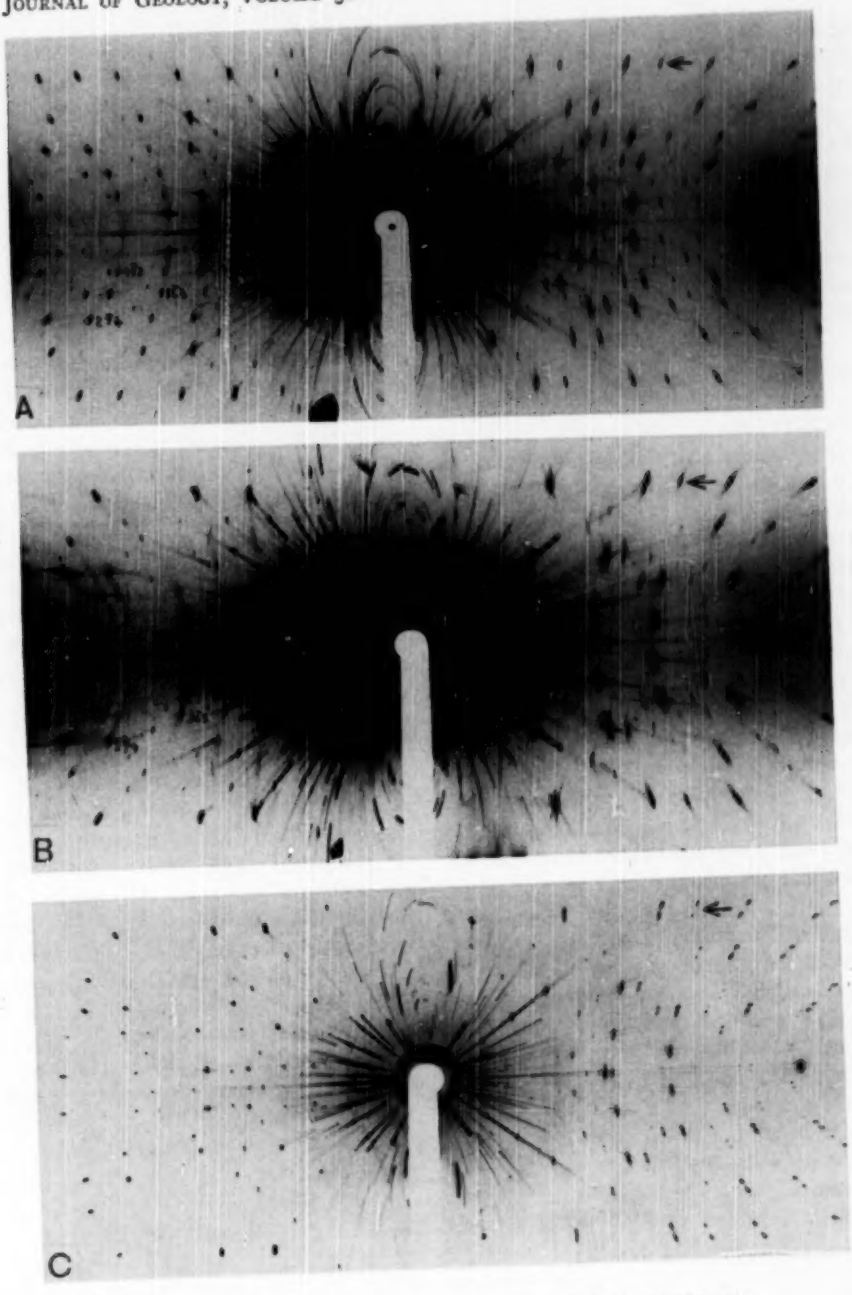
More rigorously, it can be said that the suggested imperfections (triclinic areas of albite twinning, with lattice geometry similar to that determined for microcline or for triclinic adularia) will influence the

PLATE 3

Oscillation photographs of adularia samples. Oscillation axis = *b*, Cu-radiation.

A, *B*, Photographs taken with the same crystal of a "monoclinic" adularia. The oscillation range was 21° in *A*, 10° in *B*. Note the diffuse reflections on the right side of the photographs and the sharp reflections on the left. The X-ray photograph in *B* was deliberately overexposed to show more clearly this phenomenon, which is described in the text. Note also the asymmetry of the (661) spot indicated by an arrow. Compare this with the two spots, also indicated, on photograph *C*.

C, An albite twin of triclinic adularia (same sample as shown in pl. 2, *C*). The oscillation range is 10°. Because of the geometrical relations between albite twinning and the lattice angles for triclinic adularia as determined in this investigation, the reflections of planes close to the [401] zone coincide, whereas others are separately resolved. This phenomenon is seen by comparison of the left with the right side of the photograph. The diffuse streaks in *B* correspond to the doubled spots in *C*. Note that the two spots indicated by an arrow [the upper spot is (661) of one twin, the lower is ($66\bar{1}$) of the other twin] have different intensities. The intensity difference as a function of the Bragg angle is the same as shown in the corresponding diffuse streak in *B*. (The asymmetry of the diffuse streak in *B* is *not* due to oscillation range limits, although this effect is seen in other diffuse streaks, for example, the one immediately to the left of that indicated by an arrow.) Photographs of microcline show that the intensity difference between (661) and ($66\bar{1}$) is even stronger, as the ($66\bar{1}$) reflection does not even appear in equivalently exposed photographs. The small written-in indices on *A* and *B* should be neglected.



X-ray oscillation photographs of adularia ("monoclinic" and "triclinic")

X-ray effects in such a manner that the more the reflecting plane deviates from the [401] zone, the greater will be the effect of the imperfections. Precisely this behavior is illustrated by the X-ray photographs of "monoclinic" adularia (fig. 6).

DIFFUSE-STREAK INTENSITIES

The qualitative agreement between the assumption (discussed in the foregoing section) and the actual X-ray results may be extended by some remarks on the distribution of intensity along the diffuse streaks. Of the approximately six hundred spots measured, about 20 per cent showed differences in intensity above and below the main spot. That is, the diffuse portion with a Bragg angle greater than the main spot was in these cases more or less intense than the diffuse portion with a Bragg angle smaller than the main spot. This is illustrated in plate 3, *B* (see arrow), where such a diffuse streak with an especially pronounced asymmetry can be seen.

As discussed in the first two sections, microcline as well as triclinic adularia shows differences in (*hkl*) and ($h\bar{k}l$) intensities. This is shown again in plate 3, *C*, the photograph being one taken of the same triclinic adularia albite twin seen earlier in plate 2, *C*, as a precession photograph. The photograph in plate 3, *C*, is an oscillation picture, Cu-radiation, with 10° oscillation of the *b*-axis. Because it is an albite twin, the (*hkl*) spots, which would be single in monoclinic adularia, will (except in the [401] zone) be split into two spots (*hkl*)—($h\bar{k}l$) in triclinic adularia. Plate 3, *B* and *C*, illustrates this effect. The arrow in plate 3, *B*, points to *one* (*hkl*) reflection in "monoclinic" adularia, and in plate 3, *C*, the arrow points to *two* reflections (*hkl*)—($h\bar{k}l$) in triclinic adularia. (The same oscillation range relative to the *A*-axis was used in both photographs.) One can observe that the two spots (*hkl*)—($h\bar{k}l$) marked with the arrow in plate 3, *C*, have different intensities; the intensity of (*hkl*), which has the larger Bragg angle, is greater than that of ($h\bar{k}l$), with the smaller Bragg angle. Analogously, the diffuse streak (marked with an arrow in pl. 3, *B*) has greater intensity on the side with the larger Bragg angle than on the side with the smaller angle.

The intensity differences between (*hkl*) and ($h\bar{k}l$) are, as we have seen in the section on triclinic adularia, qualitatively the same in microcline and triclinic adularia, although quantitatively greater in microcline. Therefore, the discussion in the foregoing paragraph, which compares the intensities of "monoclinic" adularia and triclinic adularia streaks and spots, is also valid in the case of "monoclinic" adularia versus microcline, with an even more pronounced difference. Inasmuch as the writer's experimental data on microcline is, to date, more complete than that on triclinic adularia, the observations to follow are based on photographs of microcline, keeping in mind that the conclusions to be drawn, qualitative as they are, may hold for triclinic adularia as well as for microcline.

It has been found that all diffuse streaks that show asymmetrical intensity correspond to planes which in microcline produce distinct intensity differences between (*hkl*) and ($h\bar{k}l$) spots. Conversely, whenever the (*hkl*)—($h\bar{k}l$) pairs in microcline showed an intensity ratio of 3 or larger, asymmetrical streaks with the corresponding indices were observable in "monoclinic" adularia. The relation of Bragg angle to intensity as discussed above (pl. 3, *B*, *C*) is valid for all these spots.

On the whole, it appears that the

greater the asymmetry of the streaks in "monoclinic" adularia, the greater is the intensity difference in the corresponding (hkl)—$(\bar{h}kl)$ spots in microcline. An analogous relation holds for "monoclinic" adularia and triclinic adularia, at least as far as could be determined from those photographs of triclinic adularia that could be used for this purpose. Neither case, however, showed quantitative relationships.

TABLE 7

RELATIONS BETWEEN THE INTENSITY DIFFERENCE OF (hkl) AND $(\bar{h}kl)$ IN MICROCLINE AND THE ASYMMETRY OF THE DIFFUSE STREAKS IN "MONOCLINIC" ADULARIA

(hkl)	$R(hkl)$	Direction of Asymmetry	(hkl)	$R(hkl)$	Direction of Asymmetry
716	$\frac{1}{4}$	+	425	4	−
515	$\frac{1}{10}$	+	113	4	−
625	4	−	823	3	−
645	4	−	626	$\frac{1}{4}$	+
735	$\frac{1}{4}$	+	535	2	−
715	$\frac{1}{10}$	+	735	$\frac{1}{4}$	+
514	$\frac{1}{3}$	+	241	2	−
825	12	−	551	2	−
734	$\frac{1}{4}$	+	223	$\frac{1}{4}$	+
333	3	−	243	2	−
513	$\frac{1}{3}$	+	133	$\frac{1}{4}$	+
553	$\frac{1}{3}$	+	643	$\frac{1}{4}$	+
221	6	−	823	3	−
311	$\frac{1}{12}$	+	314	$\frac{1}{4}$	+
531	$\frac{1}{6}$	+	114	4	−
621	3	−	241	$\frac{1}{4}$	+
330	$\frac{1}{3}$	+	711	4	−
510	$\frac{1}{3}$	+	225	$\frac{1}{4}$	+
441	$\frac{1}{3}$	+	025	$\frac{1}{3}$	+
621	8	−	115	16	−
711	4	−	331	4	−
332	8	−	621	8	−
352	8	−	222	4	−
422	4	−	440	$\frac{1}{3}$	+
442	3	−	336	8	−
513	6	−			

NOTE TO TABLE 7

Inasmuch as (hkl) and $(\bar{h}\bar{k}\bar{l})$ have the same θ (Bragg angle), it is always possible, in comparison of the pairs (hkl)—$(\bar{h}kl)$, to choose the indices (in the triclinic case) in such a way that $\theta(hkl) < \theta(\bar{h}kl)$. The indices are here chosen in this fashion. Let the intensity ratio $(hkl) : (\bar{h}kl)$ be called R. A plus (+) sign indicates that the asymmetry of the streak in "monoclinic" adularia is greater on the side of greater θ (relative to the main spot). A minus (−) sign indicates the reverse relation. Thus the relationship between the intensities of microcline and the asymmetry of the diffuse streaks in "monoclinic" adularia becomes obvious: an R-value less than unity is always accompanied by a (+); a value greater than unity, by a (−) sign.

This relationship is based on visual intensity estimations of all streaks appearing on oscillation photographs of "monoclinic" adularia with b as the oscillation axis (Fe-radiation, 10° oscillation, with an exposure time of 10 hours at 10 Ma, 45 kv). In table 7 are listed the indices of those streaks that show asymmetry of intensity. This table also includes the intensity differences of the corresponding (hkl)—$(\bar{h}kl)$ microcline spots.

It is the writer's opinion that a sufficient number of measurements have been made to justify the conclusion that the relation between the asymmetry of the streaks in "monoclinic" adularia and the intensity differences in corresponding (hkl) and $(\bar{h}kl)$ spots in microcline (and in triclinic adularia) is not a chance relationship. It supports the statement earlier made (see section on relation of [401] zone, etc.) that triclinic areas are present in the samples of "monoclinic" adularia investigated and that these triclinic areas have an atomic arrangement similar to that in microcline or in triclinic adularia. These regions are present as thin lamellae with albite and pericline twin relations. The pericline twinning was but briefly mentioned and was not discussed in detail because the diffuse phenomena produced in these crystals by pericline twinning are less striking than those due to the albite twinning. It should be mentioned, however, that planes (0k0), for example, show diffuseness in a direction near [401] which corresponds to the direction in reciprocal space that connects the points (hkl)—$(\bar{h}kl)$ of a pericline twin. Figure 8 shows how the triclinic

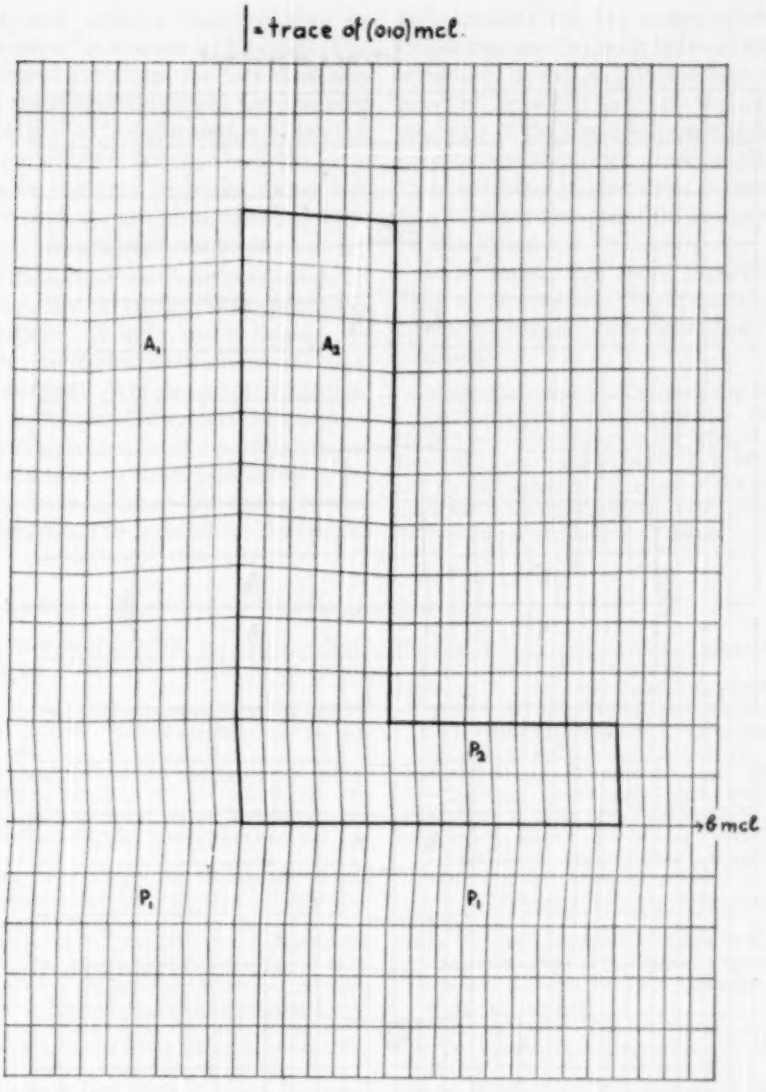


FIG. 8A

FIG. 8.—Two models illustrating triclinic areas twinned after the albite (A_1, A_2) and pericline (P_1, P_2) laws, embedded in a monoclinic lattice. These schematic drawings illustrate the relations in a plane approximately perpendicular to the *c*-axis. The greater part of the strain produced by the transformation will be at the ends of the twin lamellae, indicated on the drawing by several lines. Continued growth in the direction of the twin plane would be expected under these conditions, and elongated lamellae are indeed the rule.

Notice in *B* that A_1 lies on the left side in the upper albite position, and on the right side in the lower. This crossing-over of lamellae is commonly observed in thin sections of microcline (Rosenbusch and Mügge, 1927).

As in fig. 4, deviation from monoclinic symmetry is exaggerated for clarity.

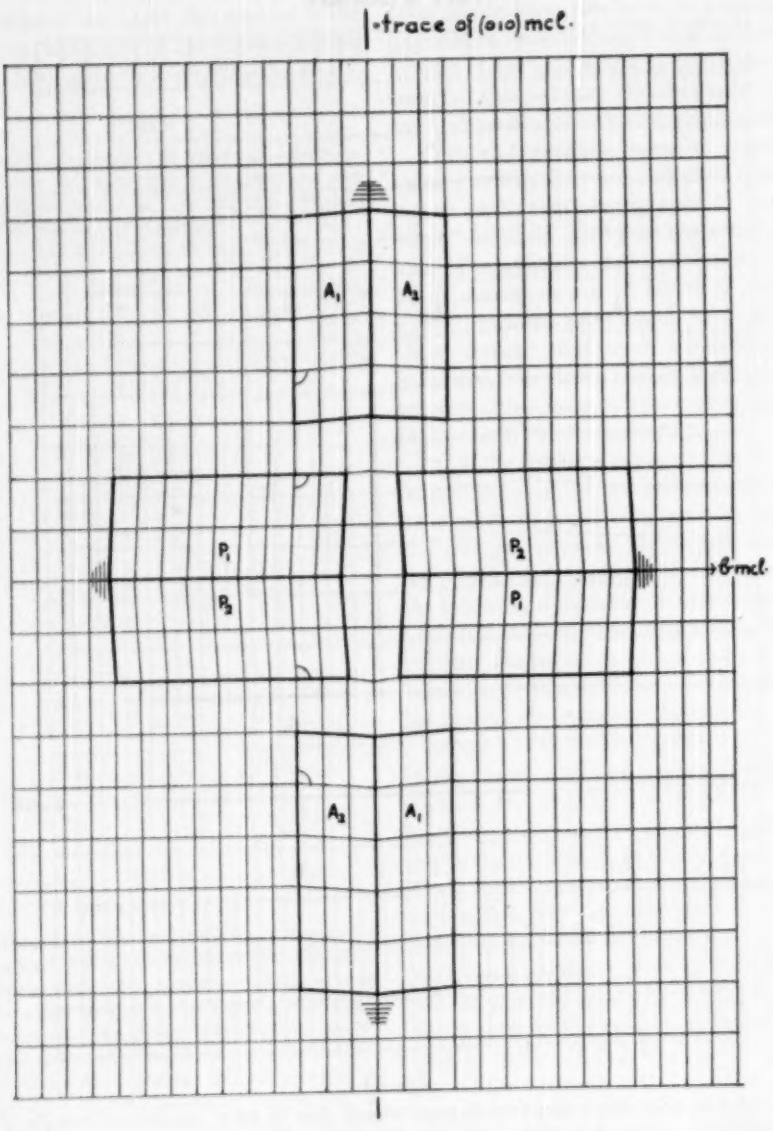


FIG. 8*B*

albite and pericline lamellae may be "embedded" in a monoclinic lattice. This figure well illustrates the fact that only small structural distortions are produced by this partial transformation, inasmuch as the albite twin lamellae have the (010) plane in common with the monoclinic lattice and the pericline lamellae have the [010] direction in common.

To date, it has not been possible to decide whether the triclinic areas present in monoclinic adularia most closely resemble microcline or triclinic adularia. The intensity data do not fit quantitatively either possibility, but with respect to the direction of intensity variation (as shown in table 7) either possibility could qualitatively account for the observed photographs. Two possibilities that might well account for the quantitative discrepancies are: (1) both types of triclinic structure, as well as intermediate states, may be present, and (2) the relative amounts of the two albite twin positions may be variable from place to place or from specimen to specimen. A combination of these two possibilities must also be considered.

It is hoped that more experimental work on "monoclinic" feldspars from different occurrences may help answer this question.

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HIGH-TEMPERATURE ALBITE AND CONTIGUOUS FELDSPARS[1]

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ABSTRACT

Synthetic albite is found to have X-ray and optical properties different than those of the very pure natural albites from pegmatites, but these natural albites can, by heating, be converted into a form identical with the synthetic product. High- and low-temperature modifications of albite are thus indicated. The inversion temperature is believed to be in the neighborhood of 700° C. Optical and X-ray studies show that the inversion extends into the plagioclase series approximately to An_{15}. These high-temperature plagioclases are common as phenocrysts in extrusive rocks but have not been found in plutonic rocks.

INTRODUCTION

The problem of the polymorphic modifications of the alkali feldspars is particularly vexing to the mineralogist. In spite of the extensive and intensive studies, there is little agreement among mineralogists as to the number of modifications or their stability ranges. The soda-rich feldspar which will be examined here is reported to have as many as four modifications (Alling, 1936). Properties that are characteristic and distinctive of the various forms are not given; the modifications being arrived at largely by analogy with the potash-rich compositions.

The existence of a high-temperature monoclinic form has been suspected by many investigators because experimental studies indicate a complete series of solid solutions at high temperatures between the monoclinic potash feldspar and the soda feldspar. During an investigation of the stability relations in the system soda feldspar–potash feldspar–water, it was discovered that the soda feldspar, being synthesized, had X-ray and optical properties notably different from those of natural albite in pegmatites and granites. As will be shown below, this synthetic soda feldspar is different from all previously suggested modifications of albite but is like the soda-rich plagioclase phenocrysts from certain extrusive rocks.

[1] Manuscript received April 18, 1950. Presented at the November, 1949, Annual Meeting of the Geological Society of America.

PREVIOUS INVESTIGATIONS

Dana (1906) was probably the first to suggest a second form of albite. He reported an "abnormal" albite from the Azores which had been studied by Fouqué. The analysis given by Fouqué (1894) indicates the composition $Ab_{86}Or_9An_5$. The optic axial angle $2V = 45°$ (−), and the extinction on (010) is 9°. The indices of refraction determined by the prism method are given as: $n_\alpha = 1.5234$, $n_\beta = 1.5294$, and $n_\gamma = 1.5305$. The optical data, which are probably more reliable than the chemical, indicate a composition near $Ab_{70}Or_{30}$ and suggest that this feldspar is anorthoclase rather than albite.

Barbier and Proust (1908) reported a monoclinic soda feldspar which was later named "barbierite" by Schaller (1911). Other investigators pointed out that the chemical analysis was not reliable and the optical measurements inadequate (Mäkinen, 1917; Alling, 1923, p. 297; Barth, 1929, 1931). Barth (1929) reported soda feldspar from Seiland which

was optically monoclinic, but it was shown by X-ray studies to be made up of submicroscopically twinned triclinic units. It must be concluded that the natural soda feldspar has been found only in the triclinic form. Barth (1931) writes: "No data whatever have been set forth so far indicating the existence of a monoclinic modification," but Merwin has shown that an abrupt change in the birefringence of natural albite takes place around 900° C.

Merwin (1911) reported that both orthoclase and albite appear in two forms, with a transition temperature about 900°. The transition was reported to be very sluggish even at 1,100°. Properties distinctive of the two forms of soda feldspar were not given.

Winchell (1925) suggests three forms of soda feldspar: barbierite, albite, and analbite. As mentioned above, barbierite has been discredited, and the name "analbite" was suggested for the "abnormal albite" of Dana, already discussed. However, analbite was placed at the bottom of the temperature scale despite the fact that the analyses given are of feldspar phenocrysts from lavas. The situation is further confused by the fourth form suggested by Alling (clevelandite), which is apparently only a habit modification of normal albite.

The four modifications of soda feldspar suggested by various investigators can thus be reduced to one, the ordinary albite of pegmatites and granites. The only evidence for a second form lies in the heating experiments of various workers, but in no case are properties given for distinguishing the high-temperature modification.

PRESENT EXPERIMENTAL STUDIES

Soda feldspar synthesized by crystallization of a glass of albite composition has X-ray and optical properties somewhat different from those of ordinary albite in granites and pegmatites. On prolonged heating at temperatures a few degrees below the melting point, natural albite changes to a form essentially identical with the synthetic material. The time necessary to bring about this change varies greatly from one specimen to another; for example, a very pure albite from Amelia County, Virginia, changes completely to the high-temperature modification on heating at 1,050° for 10 days, whereas another pure albite from Varuträsk, Sweden, changes only partially in 3 weeks, and albite from Avondale, Pennsylvania, changes very little in that time. The completeness of the change is estimated from optical examination and from X-ray spectrograms, which reveal a change of certain characteristic spacings.

Partially inverted material commonly shows under the microscope cleavage fragments with a rounded core of slightly different optical properties. It is presumed that this core represents uninverted material. The inversion evidently begins at the surface and gradually migrates inward.

The high-temperature modification has been synthesized from glass under pressure of water or water vapor at temperatures from 250° to 1,000°. It is believed that the crystals grown at the lower temperatures have formed metastably, a common behavior with many silicates. Because glasses are predisposed to give high-temperature modifications, other initial materials, such as a mixture of quartz, corundum, and sodium silicate, were tried, but only the high-temperature form was obtained. Many runs were made, using various low-temperature albites as seeds, but no evidence of growth of low albite on the seeds was dis-

covered. Again different initial materials were tried with the seeds, but with the same result.

It is apparent that the only remaining way to obtain information on the temperature of the high-low-albite inversion is by heating the low-temperature modification at successively lower and lower temperatures in the attempt to find a temperature below which the inversion will not take place. This has been carried out with sodium disilicate solution in water under pressure as a flux to hasten the transformation. At 775° Amelia albite changed to the high modification in 3 days, and at 720° the change took place in 2 weeks, whereas at 675° no evidence of change was found on heating for 3 weeks. These results indicate that the inversion may be near 700°. However, there is no assurance that longer heating at lower temperatures will not cause transformation, and a definite value for the inversion temperature cannot be regarded as established.

X-RAY DIFFRACTION PATTERNS OF HIGH AND LOW ALBITE

X-ray spectrograms have been made with a Phillips X-ray spectrometer. Samples to be X-rayed were prepared by mounting on glass slides approximately 10 mg. of finely ground material made into a sludge with acetone, to which approximately 1 per cent clear lacquer had been added.

Figure 1 shows X-ray charts of natural low-temperature albite, the same material converted to the high modification by heating dry in air, and synthetic material prepared by crystallizing glass of the composition of albite in the presence of water vapor at 800° and 1,000 kg/cm² pressure. Spacings and intensities are tabulated for various albites in table 1. Measurements of 2θ are reproducible to somewhat better than ± 0.05.

The 3.01 Å spacing is perhaps the most diagnostic of the high-temperature forms. Low albite has no comparable spacing. However, nearly all the reflections shift somewhat from the low to the high form, and there is little difficulty in distinguishing the two forms by means of powder patterns.

OPTICAL PROPERTIES OF HIGH AND LOW ALBITE

Optical properties of the two modifications are given in table 2. The most striking difference is in $2V$, which is $+83°$ in the low form and $-45°$ in the high. Twinning in the synthetic material makes it very difficult to measure $2V$ accurately, and the material which has been inverted from the low form by heating has a variable $2V$ ($45°$–$55°$). The best value is probably near 45°, as is indicated by natural high-temperature sodic plagioclases when their values are projected to pure albite (fig. 4). Synthetic and heated natural albite have significantly lower indices of refraction than the natural low-temperature material. Extinction angles on (010) cleavage measured against (001) are 20° for low albite and 9° for the high form. Maximum extinction in the zone $\perp$ (010) is 19° for low albite and approximately 30° for high albite.

High albite appears to have twinning laws very similar to the low form, although the synthetic material is not of a quality that permits accurate optical studies of the twins. Carlsbad and albite twins are certainly represented in the high modification. Heated low-temperature material apparently develops twinning during the inversion. Some twins having the composition plane parallel to

(010), and others parallel to (001) have been noted. It is believed that a careful study of the various twins in the synthetic, the natural high-temperature, and the inverted low-temperature material will give much valuable information which can be used in interpreting the history of natural plagioclases.

Changes in optical properties of low albites on inverting to the high form were studied by coating the (010) cleavage with colloidal palladium before heating and observing the changes after the X-rays indicated complete transformation to the high modification. The palladium distributes itself in small globules over the cleavage surface and there is no difficulty in recognizing the (010) plane after heat treatment. The universal stage is used to detect changes in optical properties. The (010), (001), and (110) cleavages can be recognized and plotted on the stereographic net, as well as the optical directions (fig. 2). There is one possible source of error in this type of measurement. It is not known whether the

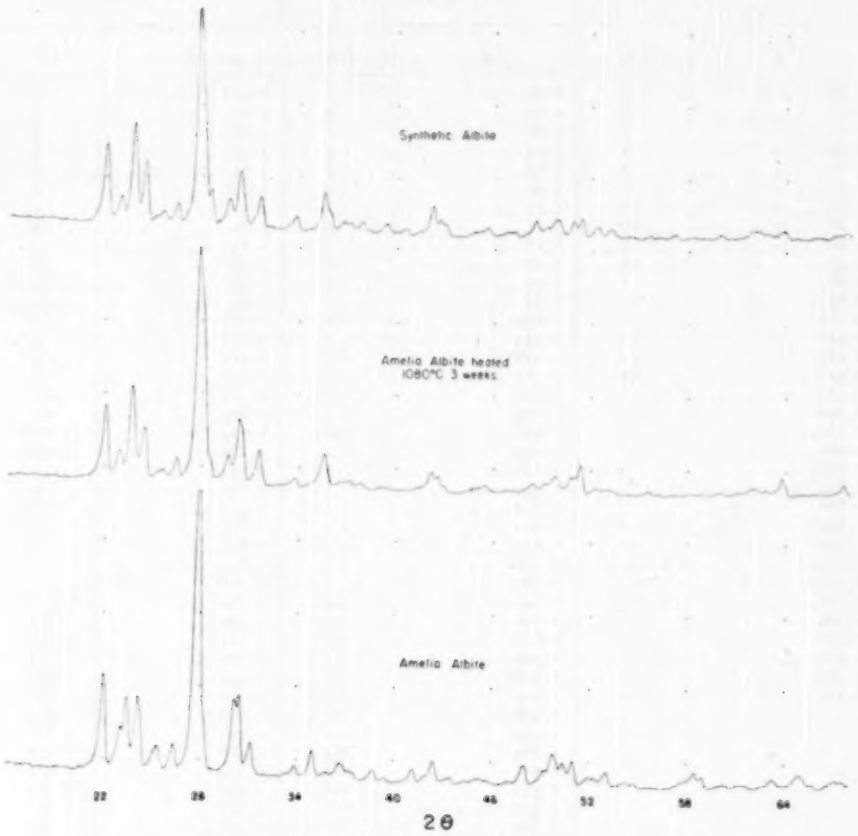


FIG. 1.—X-ray spectrometer charts of high- and low-temperature albite

cleavages being measured on the high form actually represent the cleavages of the high modification or whether they are relic cleavages of the low form. The optical orientation of the heated material agrees with that of synthetic high albite and of natural sodic plagioclase phenocrysts from rhyolites.

The existence of a high-temperature monoclinic soda feldspar has long been suspected because of the apparently complete series of solid solutions between soda feldspar and monoclinic high-temperature potash feldspar. High albite, however, is triclinic, as evidenced by the presence of albite twinning and by the

TABLE 1

SPACINGS AND INTENSITIES FOR VARIOUS ALBITES

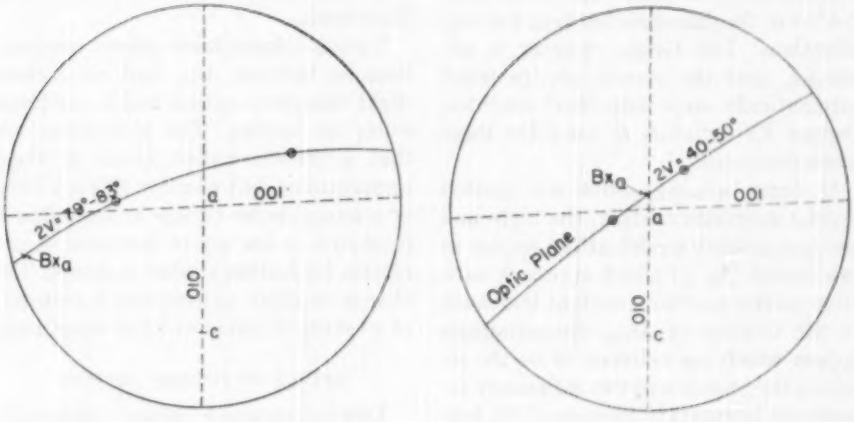
Amelia Albite		Synthetic Albite 750° C.		Heated Amelia	
d	*I/I*$_1$	*d*	*I/I*$_1$	*d*	*I/I*$_1$
6.38	1	6.39	1	6.40	1
4.02	3	4.03	3	4.03	3
3.83	2	3.88	1	3.88	1
3.79	2	3.74	4	3.75	4
3.67	3	3.64	3	3.64	3
3.51	1	3.49	1	3.49	1
3.38	1	3.37	1	3.37	1
3.20	10	3.21	10	3.21	10
2.96	2	3.13	2	3.14	2
2.93	3	3.01	1	3.02	1
2.86	1	2.95	2	2.94	3
2.64	<1	2.84	1	2.84	2
2.562	1	2.650	<1	2.655	<1
2.448	<1	2.524	1	2.525	1
2.412	<1	2.448	<1	2.455	<1
2.325	<1	2.422	<1	2.421	<1
2.280	<1	2.377	<1	2.377	<1
2.190	<1	2.295	<1	2.283	<1
2.126	1	2.220	<1	2.222	<1
2.081	<1	2.143	1	2.188	<1
1.893	1	2.122	1	2.142	1
1.832	1	1.990	<1	2.124	1
1.808	1	1.874	1	1.992	<1
1.790	1	1.842	<1	1.878	<1
1.755	<1	1.829	<1	1.842	1
1.723	<1	1.796	1	1.833	1
1.676	<1	1.780	1	1.798	1
1.576	<1	1.747	<1	1.781	1
1.564	<1	1.723	<1	1.749	<1
1.535	<1	1.613	<1	1.727	<1
1.507	<1	1.542	<1	1.620	<1
1.468	<1	1.500	<1	1.495	<1
1.437	<1	1.490	<1	1.480	<1
		1.458	<1	1.462	<1
		1.394	<1	1.418	<1
		1.384	<1	1.386	<1
		1.371	<1	1.372	<1
		1.358	<1	1.357	<1
		1.326	<1	1.348	<1
		1.266	<1	1.328	<1
		1.219	<1		

extinction angles. The possibility of a triclinic → monoclinic displacive transformation in albite was investigated by differential heating curves through the range 100°–1,000°, with no evidence of a heat effect. Also, G. L. Davis, of this laboratory, investigated the X-ray powder spectra of high albite at elevated temperatures and reported (unpublished): "A sample of synthetic high albite was x-rayed at intervals of 50° from room temperature to 1,050°. Careful examination of the powder spectra recorded between $2\theta = 20°–33°$ showed no evidence of an inversion, although preliminary records at elevated temperatures indicated such a possibility." It is concluded that the high-temperature form is triclinic and that a monoclinic modification is not the answer to the dilemma. Presumably the possibility still exists of a nonquenchable, order-disorder transformation, which would result in a gradual change to monoclinic symmetry with increasing temperature.

TABLE 2

OPTICAL PROPERTIES OF THE TWO ALBITE MODIFICATIONS

ALBITE	n_α	n_β	n_γ	$2V_\alpha$	EXTINCTION		
					On (010)	On (001)	Max. in Zone ⊥ (010)
Amelia	1.529 (2)	1.533	1.539 (2)	97	20	3	19
Heated Amelia	1.527 (3)	1.532	1.534 (4)	45–55	.....	.....	27–36
Synthetic	1.527 (2)	1.532	1.534 (1)	~45	9	0–2	25–38



A B

FIG. 2.—Stereograms of low (*A*) and high (*B*) albite. Bx_a = acute bisectrix

EFFECT OF ANORTHITE CONTENT ON OPTICAL AND LATTICE RELATIONS

X-ray powder spectrograms of low-temperature plagioclase are easily distinguished from those of the high form in compositions near albite; but, as the composition approaches An_{40}, it becomes increasingly difficult to establish differences (fig. 3). This similarity at An_{40} suggests that the structural change resulting in the inversion in soda-rich plagioclase is not present at An_{40} or, if present, is much subdued by the presence of 40 per cent anorthite.

The effect was studied by utilizing certain spacing shifts in the two modifications. The $(\bar{1}\bar{3}2)$ and $(1\bar{3}1)$ reflections are 1°.06 apart in low-temperature albite, and similar reflections in the high variety are 2°.03 apart. These reflections have been measured by using a Phillips X-ray spectrometer modified in such a manner that 5-minute counts are made at 0°.05 intervals throughout the region of the reflections being investigated. This permits using narrow slits, with consequent reproducibility of somewhat better than ±0°.02 in the difference between the two reflections. The Geiger counter is advanced, and the counts are recorded automatically on a strip chart recorder. Copper Kα-radiation is used for these measurements.

If these spacing shifts are plotted against anorthite content, the high- and low-temperature modifications appear as two curves (fig. 5) which approach each other as the anorthite content increases. In the vicinity of An_{35}, discontinuities appear which are believed to be the result of the presence of two intimately intergrown feldspars in the case of the low-temperature series.

This discontinuity in the low-temperature modification at An_{35} is evidenced also from optical properties of the two series. Figure 4 is based on a compilation of measurements of the optic axial angle of plagioclases. The optic axial angle, $2V$, of the high-temperature series in the soda-rich region obviously approaches the low with the addition of anorthite, and the two become nearly the same at An_{35}. The data suggest that $2V$ of the low-temperature form has a discontinuity in the neighborhood of An_{35}. Evidence for such a break is shown also by the indices of refraction (Chayes, 1950).

Natural plagioclase feldspars in the composition range An_{35}–An_{65} apparently consist of two phases in many cases. The evidence is shown in figure 5, where the vertical dashed lines indicate that two values are obtained for the angle between the $(\bar{1}\bar{3}2)$ and $(1\bar{3}1)$ reflections. Heat treatment of these inhomogeneous feldspars results in a merging of the two sets of reflections, the final value lying near the curve for the synthetic high-temperature series. Natural plagioclases may occur as either of the two types. No evidence of optical inhomogeneity was discovered.

Natural plagioclases whose compositions lie between An_{70} and An_{100} show slight changes in optical and X-ray properties on heating. The indications are that a transformation exists in these compositions, but whether this is a first- or a second-order change and whether it is related to the albite inversion or not remain for further studies to decide. The change in albite on inversion is certainly of a totally different order of magnitude.

EFFECT OF POTASH CONTENT

Low-temperature albite apparently cannot tolerate more than very small amounts of potash. Crystals which have formed side by side with potash feldspar usually contain only a few tenths of a

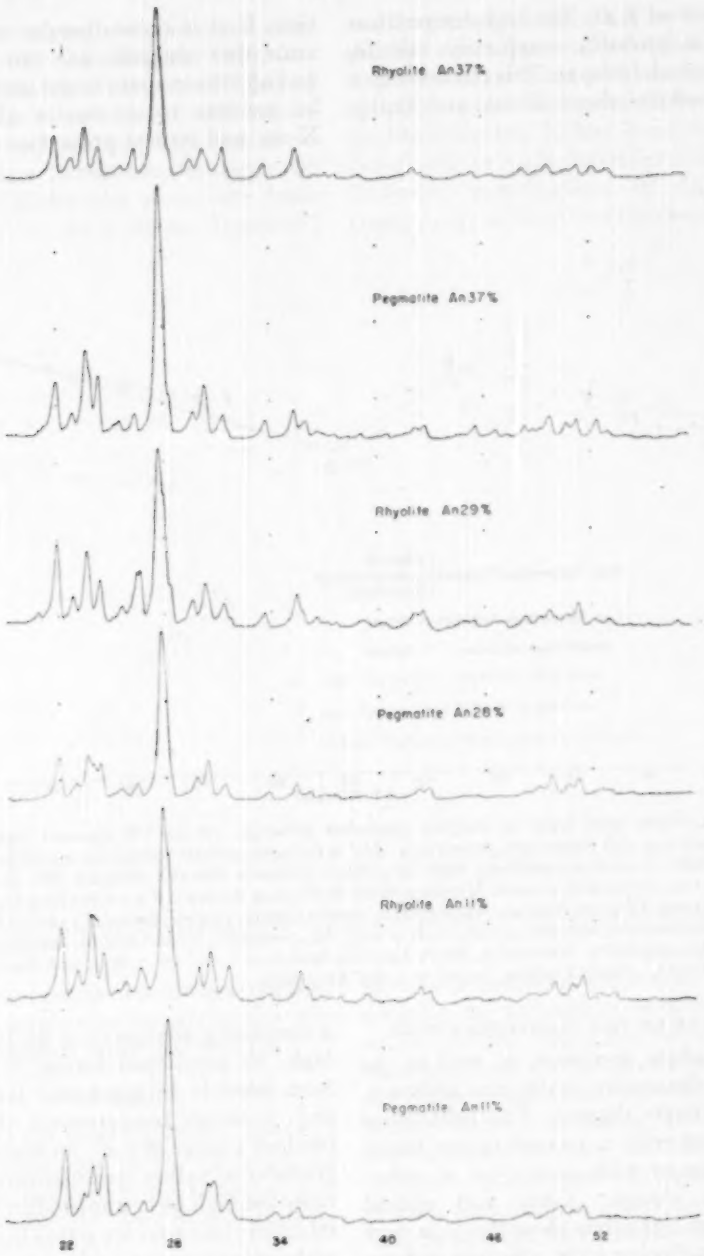


FIG. 3.—X-ray spectrometer charts of high- and low-temperature plagioclase feldspars

per cent of K_2O. The high-temperature form is probably completely miscible with potash feldspar. This relationship is discussed elsewhere (Bowen and Tuttle, 1950).

tion. If it is order-disorder and at the same time sluggish, one can expect to find all intermediate stages, and it should be possible to synthesize albite with X-ray and optical properties exhibiting

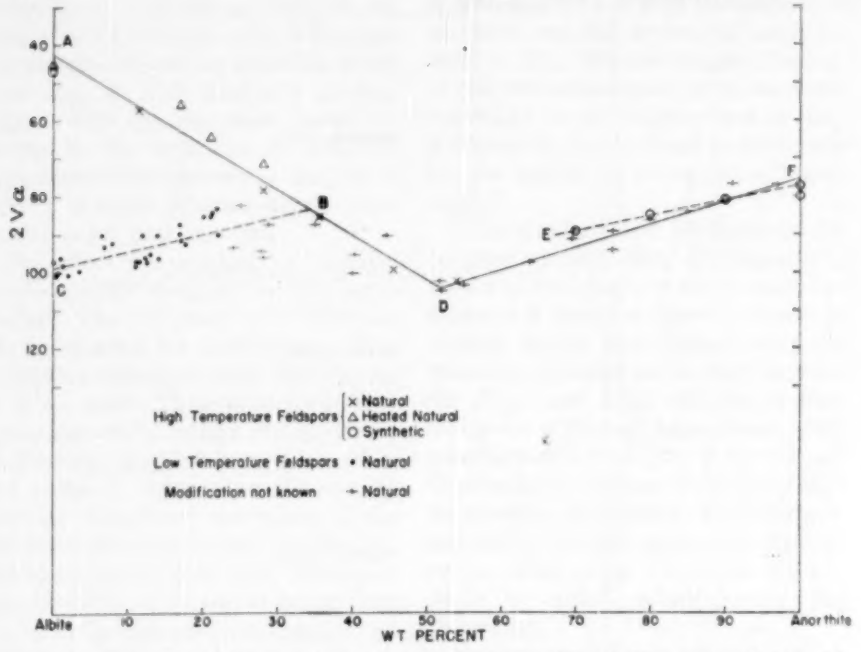


FIG. 4.—Optic axial angles of analyzed plagioclase feldspars. *AB* and *CB* represent high- and low-temperature soda-rich plagioclases, respectively. *BDF* is the most probable relation for natural low-temperature feldspars of other compositions. High-temperature synthetic lime-rich feldspars (*EF*) show a slight deviation from the natural crystals. All values except those along the line *AB* are taken from the literature. Feldspars along *AB* are by analysis: An_0 pegmatite, Amelia County, Virginia, heated at 1,080° C. for 3 weeks; An_{11} phenocrysts from rhyolite (Larsen, 1938, p. 235); An_{17} pegmatite, South Carolina, heated at 1,060° for 7 days; An_{21} pegmatite, Bakersville, North Carolina, heated at 1,060° for 7 days; and An_{28} pegmatite, Mitchell County, North Carolina, heated at 1,060° for 7 days.

NATURE OF THE TRANSFORMATIONS

The albite inversion, as well as the similar change in contiguous feldspars, is exceedingly sluggish. This behavior is consistent with a reconstructive transformation or with some type of order-disorder change. X-ray and optical studies of high albite show that it is very similar to the low form, a feature perhaps favoring an order-disorder transformation. If it is order-disorder and at the same time sluggish, one can expect to find all intermediate stages, and it should be possible to synthesize albite with X-ray and optical properties exhibiting a complete gradation from the low to the high. As mentioned before, it has not been possible to synthesize low albite, and, although temperatures of growth covered a range of 750°, no evidence of a gradational nature was discovered. In all cases the high form was produced. Natural albitic feldspars are either high or low without evidence of intermediate states.

It is concluded that the albite inver-

sion must involve some changes other than Al ⇌ Si order-disorder. The fact that both modifications are changed by the addition of anorthite in such a manner that they become very similar at An_{35} appears to support the suggestion that Al ⇌ Si disorder is not the dominant factor in the inversion. Significant differences in refractive indices (density) and the large heat of transformation[2] also suggest marked structural changes.

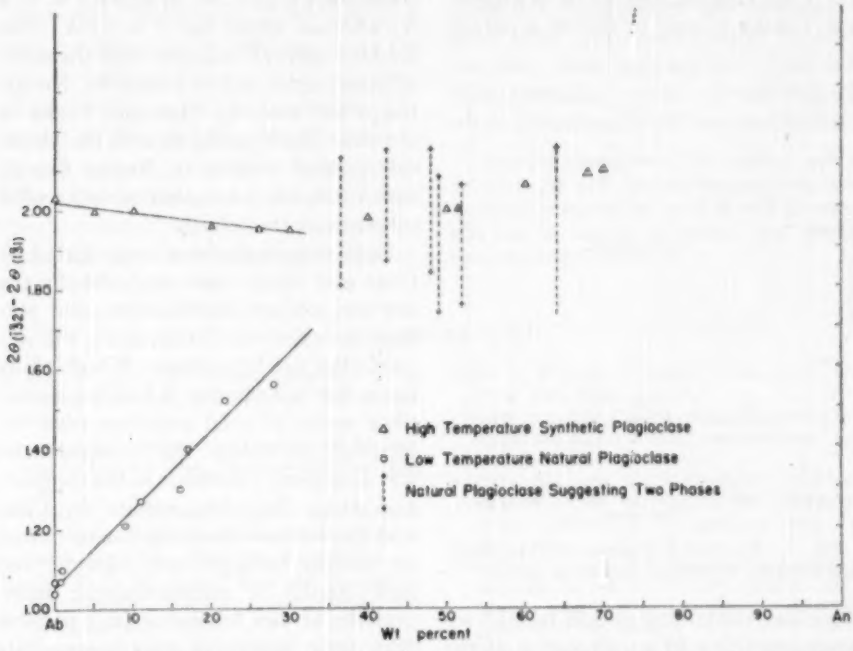


FIG. 5.—Variation, with composition, of the angle between the $(1\bar{3}2)$ and $1\bar{3}1)$ reflections in some plagioclase feldspars.

DISCUSSION

One of the most significant advances in the mineralogy of the feldspars has been the recognition by Köhler (1941) and co-workers of the existence of high- and low-temperature optics for the plagioclase series. The studies explain many anomalies which have annoyed those attempting to use the determinative charts for the feldspars. Köhler has published new charts for the high and low feldspars. However, investigations by Oftedahl (1948, p. 15) indicate that the charts may have to be revised in some cases, and he suggests the probability of transitional optics. Tertsch (1944) has heated albite from Mariupol and found changes in optics barely greater than the limit of error of measurement, at least in the values of Köhler angles. Actually, the difference of optical orientation between high and low albite is very marked, and it may, therefore, be doubted that Tertsch really had high albite.

Unusual optics have been reported by

[2] F. C. Kracek, of this laboratory, has determined the heat of transformation of albite to be approximately 9 cal/gm.

petrographers for plagioclases in the An_{0-10} composition range. For example, Oftedahl (1948, p. 34) writes: "All the physical properties suit fairly well with those of an oligoclase (15–20 An), with one exception: The axial angles are definitely too low, 62° instead of 80–90°." Again Oftedahl (1948, p. 20) writes: ". . . Then an axial angle of 68° is anomalous. I think it may be due to a potash

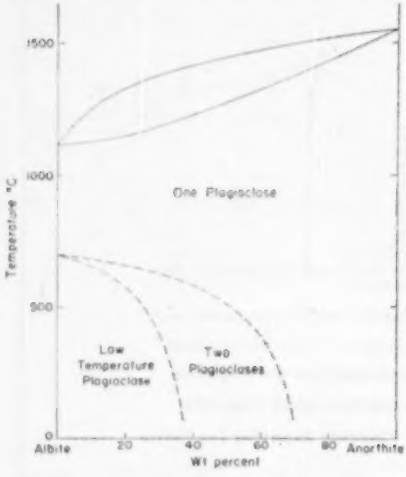


FIG. 6.—Equilibrium diagram for the plagioclase feldspars. Subsolidus relations tentative.

oligoclase, containing potash in solid solution, resulting in an alteration of the axial angle, but without influence on the other optical data." Such values may be expected in high-temperature plagioclase. Larsen (1938) reported a plagioclase containing 11 per cent anorthite and found refractive indices which necessitate a negative sign. Examination of this material, kindly provided by Dr. Larsen, reveals X-ray and optical properties of the high-temperature series (i.e., $2V = 58°(-)$; see also fig. 4).

S. H. Chao and W. H. Taylor (1940) have suggested that the plagioclase feldspars may consist of two separate isomorphous series, one with the albite structure extending to An_{22+} and the other with anorthite structure extending from An_{68-80} to An_{100} and a series of intermediate plagioclases consisting of a mixture of these two occurring as sheets parallel to (001), each sheet being a few unit cells in thickness. Anorthite-rich plagioclases have a unit cell in which $c = 14.2$ Å, whereas albite has $c = 7.2$ Å. This doubled unit cell indicates that the solid-solution series is not complete. No attempt was made by Chao and Taylor to reconcile this hypothesis with the physical-chemical results of Bowen (1913), which indicate a complete series of solid solutions at the solidus.

Soda-rich plagioclases investigated by Chao and Taylor were undoubtedly the low-temperature modification, and perhaps the existence of high albite will reconcile the two hypotheses. If high albite has a unit cell capable of forming a complete series of solid solutions with the anorthite structure, the inconsistencies will disappear. Feldspars in the composition range An_{35}–An_{65} studied by Chao and Taylor were probably changed from an initially homogeneous character and now consist of submicroscopic intergrowths of two feldspars. Is it possible that their picture of this intermediate group would be altered if homogeneous feldspar were studied in place of the probably unmixed material?

If the temperature of the albite inversion is near 700°, as is suggested by the present study, one would expect to find plagioclase crystals with cores of high-temperature feldspar and rims of the low form in some extrusive rocks. The presence of such feldspars in plutonic rocks would be decisive evidence for a magmatic history of these rocks.

However, it is possible that the high form cannot survive the excessively long

period of exposure to a moderately elevated temperature which is characteristic of plutonic conditions; but, at the same time, it is not improbable that the high form will have a "memory" of its former state, that is, it will show characters which permit ascertaining whether or not it grew as the high modification. For example, the crystal habit of the high form may be significantly different from the low, or perhaps the optics or twin laws of the two forms will be diagnostic. The nature of the twinning should certainly be different when the crystals are grown below the inversion, as compared with that developed when they are inverted from the high-temperature modification.

In figure 6 is presented an equilibrium diagram of the plagioclase feldspars which includes an interpretation of the relations at subsolidus temperatures. Albite is assigned the definite inversion temperature indicated by our studies. The inversion is lowered by solid solution of anorthite in the manner indicated. Lime-rich plagioclases are assumed to show only continuous change (Al $\rightleftharpoons$ Si disorder) with temperature. This relation probably exists throughout the whole area designated "one plagioclase."

ACKNOWLEDGMENTS.—The writers are indebted to Dr. J. F. Schairer for providing many synthetic compositions used in this investigation and for making numerous "dry" heating runs on natural feldspars.

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AN X-RAY INVESTIGATION OF THE "HIGH"-"LOW" ALBITE RELATIONS[1]

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ABSTRACT

As shown in the accompanying paper by Tuttle and Bowen, two kinds of albite exist. They are distinguished by optical properties and differences in X-ray powder patterns. The generally known "normal" albite (called "low" albite in this paper) is one type, the other (here called "high" albite) can be obtained by heat treatment of "low" albite or by direct synthesis.

This investigation deals with some of the "low-high" albite relations. X-ray investigations combined with optical measurements have been carried out. The optical measurements are similar to those reported by Tuttle and Bowen.

Structural differences as revealed by the X-ray photographs are discussed. The lattice of "low" albite differs from that of "high" albite in that (010)∧(100) changes from 90°30′ to 88°15′ and (010)∧(001) from 86°20′ to 86°0′. This lattice difference is accompanied by changes in intensities of the X-ray reflections. The structural differences, as far as they are expressed as intensity differences of the X-ray reflections, are of the same order of magnitude as the differences in microcline and orthoclase.

INTRODUCTION

The present investigation is an outgrowth of the work done by Tuttle and Bowen, as reported in an accompanying paper in this issue of the *Journal*. These authors report the large optical changes that result when albite is heated for a sufficiently long time in the proper temperature range. Using the X-ray powder method, they also show that the optical changes are accompanied by structural change. Tuttle and Bowen kindly supplied the authors with much of the material used here for a combined optical and X-ray study. The X-ray work was carried out by the senior author, the optical work by the junior author.

Spencer (1930) and Barth (1931) reported that in some cases heating albite at 1,000° C. changed the $2V$ about 8°. Barber (1936) disputed these results, but Spencer (1937) confirmed his earlier results and found that the time of heat treatment is important and also that albites from different localities behave differently. In this later work he reports a change in $2V_a$ of from 100° to 35°–60° upon prolonged heating at 1,075° C. The extinction angle on (010) also changed from 18° to 8°. Köhler (1941) and his coworkers, Scholler and Tertsch, also observed some changes, as published in diagrams by van der Kaaden (1949).

Tuttle and Bowen show that all albite that they have synthesized, in a temperature range that goes as low as 300° C, is "high" albite. The synthetic crystals used in the present investigation were kindly supplied by Mr. I. Friedman, one batch being grown at 300° C., another at 450°C.

All the crystals used for the X-ray study were measured optically. These measurements (with one exception) were similar to those reported by Tuttle and Bowen on both the heated natural albite (Amelia County, Virginia) and the synthetic crystals. These optical data also agree with those of Spencer (1937).

OPTICAL STUDY

Figure 1 illustrates the orientation of unheated Amelia albite as seen in a sec-

[1] Manuscript received May 26, 1950.

tion cut normal to the *c*-axis. This is the usually accepted orientation and axial angle of albite. Figures 2 and 3 are stereograms of Amelia albite heated at 1,060° for 3 weeks (material supplied by Tuttle and Bowen). The orientation shown in figure 2 is very similar to that reported by Tuttle and Bowen in the accompanying paper. Figure 3 shows an entirely different orientation and $2V$. This measurement was made on a crystal from the same batch of heated material that produced the common orientation and $2V$ of figure 2.

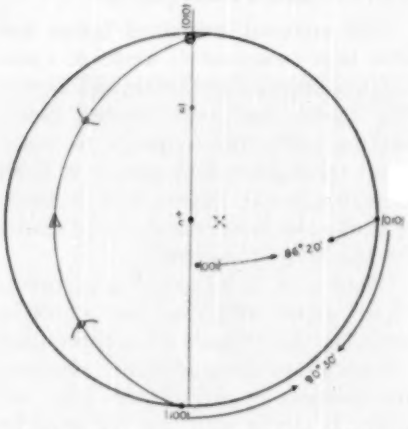


FIG. 1.—Stereogram of unheated Amelia Courthouse albite.

The hydrothermally synthesized albite here studied from the batch grown at 300° showed two distinct crystal habits, both habits being produced in the same experimental run. Some of the crystals grew as needles elongated in the direction of the *c*-axis, with (001) and (010) predominant. These needles for the most part showed fine polysynthetic albite twinning, and only a few crystals could be found with areas sufficiently free of this fine twinning to permit measurements of the optical indicatrix. The other crystals were flat tablets, nearly square in outline, lying on (010). All of the tablets were finely twinned after two laws, the pericline and one similar

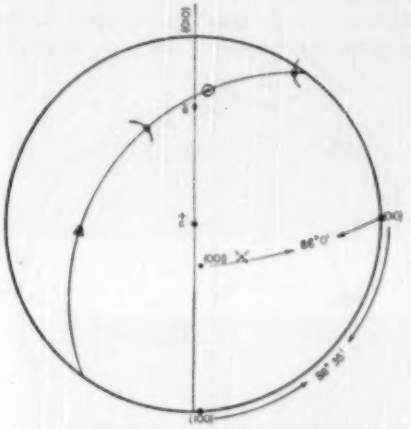


FIG. 2.—Stereogram, showing the average orientation and axial angle of two fragments of heated Amelia Courthouse albite.

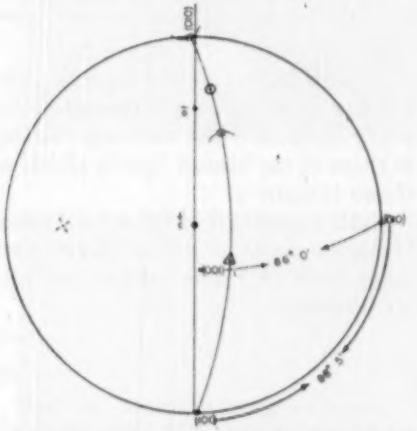


FIG. 3.—Stereogram, showing unusual optical orientation of heated Amelia Courthouse albite.

to the Carlsbad, as revealed in the X-ray study.[2] The twinning was not recognizable under the microscope. Each tablet showed an average orientation for its

[2] See explanation in the legend for pl. 2.

assemblage of twins, so that the plotting of optical orientations has no significance. It could be observed, however, that a was the acute bisectrix. On the stereogram of figure 4 are plotted the optical measurements made on eleven of

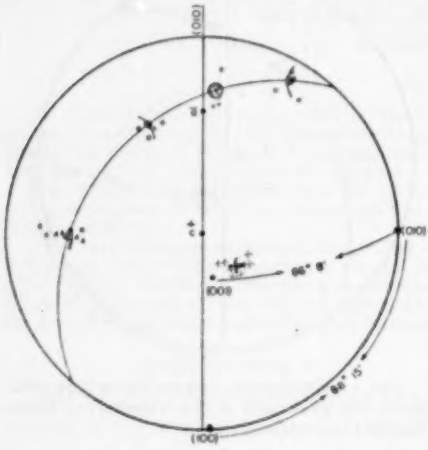


FIG. 4.—Stereogram, showing average orientation and axial angle of eleven synthetic albite needles.

the needle-like synthetic crystals. The average readings are represented by heavy symbols. These values are similar to those of the heated Amelia albite, as shown in figure 2.

Table 1 summarizes the optical measurements made on albite. X-ray data taken from the same samples are also included here.

X-RAY STUDY

The precession technique was used on all the albite samples. (See accompanying paper on potash feldspar for methods used.) All measurements here quoted have a limit of error of $\pm 5'$. Care was taken in all measurements to correlate the optical indicatrix with the chosen system of co-ordinates, as defined by a and γ. The angular values are given in table 1.

AMELIA COUNTY ALBITE

This material, examined before and after heat treatment (3 weeks at 1,060° C.), was supplied by Tuttle and Bowen. The sample had been crushed before heating, as the transformation to "high" albite takes place more readily in finely divided material. Those grains that were optically the most uniform were chosen for the X-ray photographs.

Plate 1, *A*, is a precession picture of "low" albite with c as the precession axis. The (hko) planes are here recorded. The reciprocal axes, a^* and b^*, represent the normals to the planes ($h$oo) and (oko). It can be seen that the angle between $+a^*$ and $+b^*$ is greater than 90°. [The exact value of (oko) $\wedge$ ($h$oo) is 90°30′.] Compare plate 1, *A*, with 1, *E*, the latter being a precession picture of an albite twin. The a^*-axes of the two twin individuals form an angle of 1°.

PLATE 1

Precession photographs of "low" and "high" albites with c as the precession axis. No radiation. The symbols $<90°$ and $>90°$ on the photographs refer to the angles between $+a^*$ and $+b^*$. The arrows indicate spots discussed in the text.

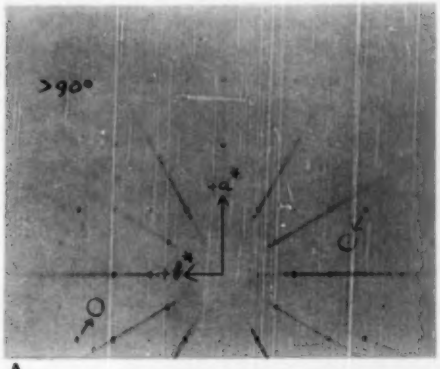
A, Photograph of "low" albite (no. 1, table 1). The observable doubling of the spots is here due to lineage.

B, Photograph of "high" albite (no 5, table 1).

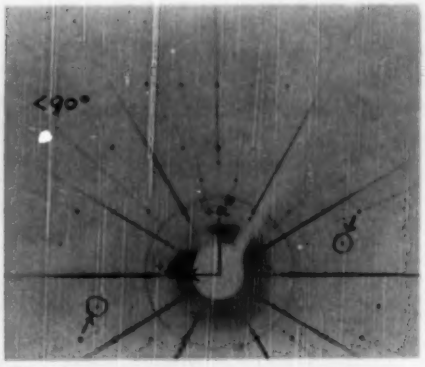
C, Photograph of "high" albite (no 2, table 1).

D, *F*, Photographs of a "high" albite twin, twinned after the albite law. Diffuse lines that can be observed between the spots are discussed in the last section of the text.

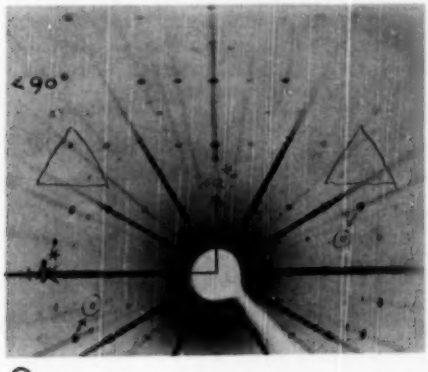
E, Photograph of a "low" albite twin (Amelia County albite), twinned after the albite law. Compare the intensities of the encircled spots with corresponding spots in *F*. Note the reverse ratio of intensities in the latter, due to the fact that the right spots of the doublet in *E* correspond to the left spots in *F*. (The value of $+a^* \wedge +b^*$ is greater than 90° in *E* and smaller than 90° in *F*.



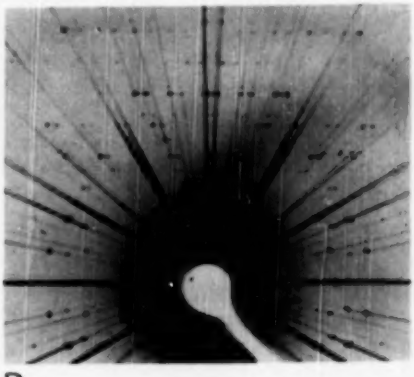
A



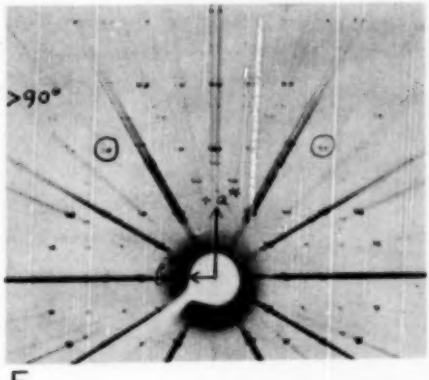
B



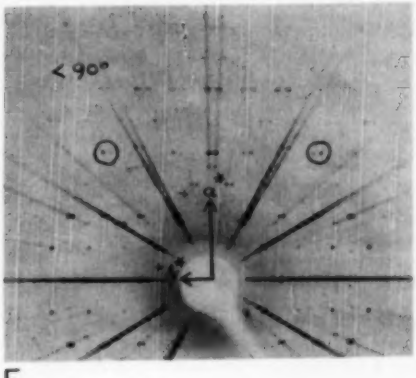
C



D



E



F

Precession photographs of low- and high-albite

Precession photographs of a special high-albite twin

Plate 1, *C*, is a precession picture of a heated grain. Compare it with 1, *A* (unheated). Note that the angle between $+a^*$ and $+b^*$ is less than 90°. [The exact value of $(o\bar{k}o) \wedge (h\bar{o}o)$ is 88°35′.] Compare plate 1, *C*, with 1, *D*, the latter being an albite twin (heated). Compare plate 1, *D* (heated), with 1, *E* (unheated), and note the difference in the angles formed by the a^*-axes. (In both cases the b^*-axes coincide because of the albite twinning.)

prepared in Morey-type bombs at 300° and 450° C. in an investigation of the albite stability field in a portion of the system Na_2O-Al_2O_3-SiO_2-H_2O. All the material formed at both temperatures was "high" albite.

TABLE 1

OPTICAL MEASUREMENTS MADE ON ALBITE

No. and Material	Fig.	Heat Treatment	Optical Data		X-ray Data			
			Bxa	*2V*	(010) ∧(100)	(010) ∧(001)	γ	α
Albite (from literature)*			γ	80°–85°	90°3′	86°24′	88°8′	94°3′
1. Albite (Amelia)	1	Unheated	γ	84°	90°30′	86°20′	87°39′	94°20′
2. Albite (Amelia)	2	1,060° C. for 3 weeks	a	64°	88°35′	86°0′	89°36′	93°44′
3. Albite (Amelia)	3	1,060° C. for 3 weeks	a	40°	88°5′	86°0′	90°9′	93°30′
4. Synthetic albite (needles)	4	Synthesized at 300° C.	a	56°±3°	88°16′	.....	.....	.....
5. Synthetic albite (needles)	....	Synthesized at 300° C.	a	56°±3°	88°15′	86° 8′	90°2′	93°20′
6. Synthetic albite (needles)	....	Synthesized at 450° C.	a	56°±3°	88°16′	86°10′	90°4′	93°26′

* Data from literature: crystallographic angles from Dana (1909); optical from DuParc and Reinhard (1924).

SYNTHETIC $NaAlSi_3O_8$

Crystals of albite suitable for single-crystal X-ray photographs were obtained from I. Friedman. These crystals were

The two habits of these crystals were briefly described in the preceding section. Plate 1, *B*, is a precession picture of one of the needle-like crystals. Comparison of this picture with 1, *C* (heated Amelia albite), shows the similarity of the angles between $+a^*$ and $+b^*$ and the similarity of spot intensities. No differences could be observed between needles grown at 300° and those at 450° C. Intensities

PLATE 2

Precession photographs of "high" albite, with *b* as the precession axis. Cu radiation. Both crystals were from the same hydrothermal run.

A, Photograph of one of the tabular crystals. The pattern shows that this platy crystal is twinned with an axis very close to *c* (i.e., very close to a^*). The indices for the spots are given in fig. 5, a schematic drawing. Precession pictures with the *c*-axis as precession axis (not presented here) show two positions related by the pericline law. Therefore, we have a "fourling" of pericline twins: $P_1 - P_2$ and $P'_1 - P'_2$. *P* and *P'* are related to each other by a twin axis that is normal to *b* (and *b'*). The twin axis thus is perpendicular to [010] and lies in the (100) plane. This corresponds to the "*X*-pericline" law, according to Berek (1924). Both twin axes are perpendicular to each other. (This might indicate that these tabular crystals had a rhombic nucleus at the beginning of crystallization. This statement is made on the basis of the reasoning presented in the accompanying paper on potash feldspar, to account for the microcline twinning.)

B, Photograph of an untwinned portion of a synthetic albite "needle."

and lattice angles are the same within the limit of experimental error (see table 1, nos. 5 and 6).

The platy albite crystals [plates parallel (010)] showed sharp edges parallel to [001]; the other two edges were ragged. Precession photographs of these crystals taken with both *c* and *b* as the precession axes showed the plates to be twinned after the pericline law and a law similar to the Carlsbad. No albite twinning is present in crystals of this habit. Compare plate 2 with figure 5.

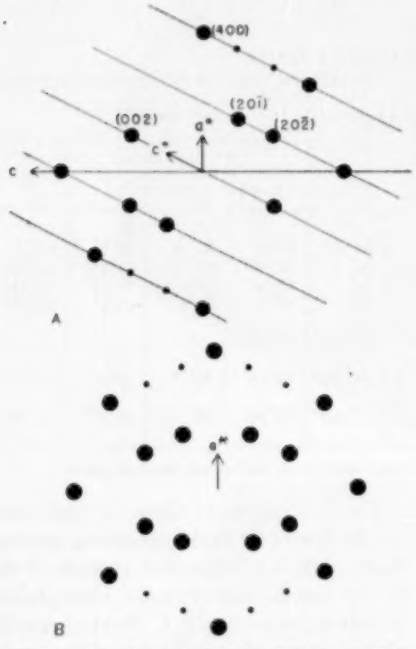


FIG. 5.—*A*, schematic drawing of the spots visible in the precession photograph (pl. 2, *B*). *B*, schematic drawing of the pattern in *A*, twinned with an axis close to *c* as twin axis. The spots have the same positions as shown in the photograph (pl. 2, *A*).

REMARKS ON THE STRUCTURAL RELATIONS BETWEEN "LOW" AND "HIGH" ALBITE

Although no quantitative evaluation of the X-ray photographs with a view toward the determination of the structure of "high" albite has been attempted, it is possible to obtain some information on the amount of structural difference between the two modifications. This was done by comparing the intensities of the two modifications in a statistical manner.

As shown in table 1, the lattice differences between the two forms are rather small. The angular differences are similar to the amount of deviation of microcline from orthoclase. (see the accompanying paper by Laves on potash feldspar). Table 2 presents the angular values for the two modifications of albite and for orthoclase and microcline. The lattice angles of "high" albite are smaller than those of the "low" form, and the table shows the angular differences of the two forms as compared to the differences between orthoclase and microcline.

As is pointed out in the accompanying paper on K-feldspar (table 4), the differences between (*hkl*) and (*hk̄l*) are associated with the change of the angles γ^* and α^* as they deviate from 90°. Tables 3 and 4 show that intensity differences of the same order of magnitude are observed between "high" and "low" albite. Plate 1, *A* ("low" albite), and 1, *B*, *C* ("high" albite), show such intensity differences, especially for the spots marked by arrows. The intensity of (1$\bar{7}$0) for "high" albite is less than (1$\bar{7}$0) for "high" albite, and the intensity of (190) for "high" albite is greater than (1$\bar{9}$0) for "high" albite, whereas in "low" albite (1$\bar{7}$0)—(1$\bar{7}$0), on the one hand, and (190)—(1$\bar{9}$0), on the other hand, have the same intensity. These same relations are found for microcline-orthoclase. Compare plate 1, *B* ("high" albite), and 1, *A* ("low" albite), with plate 1, *E*, of the accompanying paper on potash feldspar.

Table 3 lists the intensities of 55 pairs (*hk*0)—(*hk̄*0) in "low" albite and "high"

albite. These intensities were estimated from precession photographs, as described in the paper on potash feldspar. In addition to the intensities, table 3 also lists the intensity differences between (*hkl*) and ($h\bar{k}l$). These differences, compared with the intensities themselves, furnish information on the "triclinicity" of the structure. Let R be the ratio of the sum of the differences (Δ) between the intensities to the sum of the intensities. It is thus obvious that R is equal to zero in a monoclinic structure, since for monoclinic symmetry there is no intensity difference between (*hko*) and ($h\bar{k}o$). In the triclinic case the (010) plane is no longer a symmetry plane, and therefore one cannot expect the (*hko*) and ($h\bar{k}o$) intensities to have the same values. If, however, deviation from monoclinic symmetry is small, the intensity of (*hko*) will not be very different from that of ($h\bar{k}o$). Therefore, the closer a triclinic structure approaches a pseudo-monoclinic one, the smaller will be these intensity differences, and the smaller will be the ratio R.

Using the intensity values of table 3, the ratio R can be calculated for the two modifications of albite thus:

$$R\text{ ("low" albite)} = \tfrac{74}{354} = 0.21\,,$$

$$R\text{ ("high" albite)} = \tfrac{71}{285} = 0.25\,.$$

Thus it is shown (as is also apparent from the lattice geometry, table 2) that "high" albite is "more triclinic" than "low" albite. This conclusion applies for the triclinic attitudes in the zone of the *c*-axis, the zone in which this intensity method was used.

The value of R for microcline also can be obtained from the intensity data of the microcline (*hko*) planes. Comparison of the microcline value with the albite value is of interest. From table 4 of the potash feldspar paper we obtain

$$R\text{ (microcline)} = \tfrac{43}{322} = 0.13\,.$$

As expected, microcline has a structure more nearly pseudo-monoclinic than albite does, because the deviation from monoclinic symmetry (where $R = 0$) is smaller than that for albite.

It is of interest to express the deviation of the "high' albite structure from the "low" albite structure in the same fashion. As above, let

$I_{(hkl)} - I_{(h\bar{k}l)} = \Delta$ (low) for "low" albite ;

$I_{(hkl)} - I_{(h\bar{k}l)} = \Delta$ (high) for "high" albite ;

and

$$\Delta' = \Delta\text{ (high)} - \Delta\text{ (low)}\,.$$

Then R' (= sum of differences Δ' : sum of intensities of "low" albite) is a measure of the deviation of the "high" albite structure from the "low" albite struc-

TABLE 2

ANGULAR VALUES FOR THE TWO MODIFICATIONS OF ALBITE, FOR ORTHOCLASE, AND FOR MICROCLINE

Material	γ^* (010)∧(100)	Diff.	α^* (010)∧(001)	Diff.
"Low" albite	90°30′	2°15′	86°20′	20′
"High" albite	88°15′		86°00′	
Orthoclase	90°00′	2°20′	90°00′	25′
Microcline	87°40′		89°35′	

TABLE 3

INTENSITIES OF (*hk*o) PLANES OF "HIGH" AND "LOW" ALBITE*

k =	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
(8*k*o)	o o	22 3′0′ −3 +3		34 +1 34′ +1½ −½		43′ −½ 54 −1 +½		3′5 +1½ 4′5′+1 +½		10 −1 30 −3						
(7*k*o)		43 −1 43′ −½ −½		21 −1 22 −1		02 +2 12 +1 +1		1′1′ 32 −1 +1		02 +2 02 +2		02′ +2½ 0′3 +2½				
(6*k*o)	8 8			4′5′+1 56′ +1½ −½		66 7′7 −½ +½		02 +2 22 +2		00 0′0′		01′ +1½ 0′0′ +1½		02 +2 02 +2		04 +4 45 +1 +3
(5*k*o)		1′1′ 22		45 +1 4′4′ +1		3′3′ 3′3′		2′3′ +1 24 +2 −1		43′ −½ 54 −1 +½		11 02 +2 −2		2′2 −½ 3′2′ −1 +½		
(4*k*o)	8 8			56′ +1½ 56 +1 +½		2′0 −2½ 40 −4		45 +1 56 +1		1′4 +2½ 44 +2½		10 −1 20 −2		01 +1 02 +2		10 −1 40 −4 +3
(3*k*o)		5′5′ 5′5 −½ +½		03′ +3½ 05 +5		00′ +½ 01 +1		34 +1 54′ −½ +1½		00 01 +1		20′ −1½ 41 −3 +1½		0′2 +1½ 0′2 +1½		02 +2 22′ +½ +1½
(2*k*o)	o o		8 8		5′7′ +2 67 +1 +1		7′7′ 7′7 −½ +½		7′7′ 88		3′0 −3½ 40 −4		3′3′ 4′4′		0′0 −½ 0′1′ +1	
(1*k*o)		51 −4 44 −4		88 88		51 −4 50 −5 +1		5′1′ −4 4′4 −½ −3½		46 +2 55′ +½ +1½		01′ +1½ 03 +3		02 +2 03′ +3½		12 +1 44 +1
	−	−		8		8		6		6		2		5		4

* Taken from precession photographs, as in pl. 1, *B* and *A*. Mo-radiation.

NOTES TO TABLE 3

The figures 0, 1, 2, 3, 4, 5, 6, 7, 8, represent intensity values—0, 1, 2, 4, 8, 16, 32, 64, 128. The intensity values of the different modifications are arranged in the following way:

A	B
C	D
Δ′	

The *A* position is the value of (*hk*o) in "high" albite; The *B* position is the value of (*hk*o) in "high" albite; The *C* position is the value of (*hk*o) in "low" albite; The *D* position is the value of (*hk*o) in "low" albite.

The meaning of Δ′ is discussed in the text. The quantity *n*′ signifies an intensity value between *n* and (*n*+1). In some cases a difference in intensity between (*hk*o) and (*hk*o) exists. Here these intensity differences appear in extra figures in the box (Δ-values in the text). The symbol (+) signifies (*hk*o) > (*hk*o) and (−) signifies (*hk*o) < (*hk*o).

ture. Again, using the intensity data of table 3, we get

R' (relation of "high" to "low" albite)

$$= \frac{55}{354} = 0.15.$$

The $R' = 0.15$ value is quite similar to $R = 0.13$ for microcline, and one may therefore conclude that the differences in the "high" and "low" albite structures are of the same order of magnitude as the differences between the microcline and orthoclase structures. This analogy is even more apparent if one compares the *signs* and magnitudes of Δ (microcline) and $\Delta' = \Delta$ ("high" albite) $- \Delta$ ("low" albite). Table 4 lists these data. The Δ-values for those (hkl) spots that show large intensity differences between (hkl) and $(h\bar{k}l)$ in microcline [for example, (820), (480), (370), (170), (190)] are of the same magnitude and sign in the "high" and "low" albite comparison.

The statistical method here used is somewhat crude and could be refined in several respects. Refinement, however, was not deemed necessary for present purposes.

TABLE 4

INTENSITY DIFFERENCES Δ AND Δ'*

	h														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
(8k0)		+3 +3		.... $-\frac{1}{2}$		+1 +$\frac{1}{2}$		+1 +$\frac{1}{2}$							
(7k0)	.... $-\frac{1}{2}$		−1 −1		.... +1		.... +1				+2 ?				
(6k0)		.... $-\frac{1}{2}$		.... +$\frac{1}{2}$		+1 +2				.... +1$\frac{1}{2}$				+3 +3	
(5k0)	+1		.... +1				.... −1		.... +$\frac{1}{2}$		.... −2		.... +$\frac{1}{2}$		
(4k0)		.... +$\frac{1}{2}$		+$\frac{1}{2}$				+4 +2$\frac{1}{2}$		−1				+$\frac{1}{2}$ +3	
(3k0)	+$\frac{1}{2}$ +$\frac{1}{2}$		+$\frac{1}{2}$				+3 +1$\frac{1}{2}$		+4 ?		.... +1$\frac{1}{2}$		−1	.... +1$\frac{1}{2}$	
(2k0)				+$\frac{1}{2}$ +1		+1 +$\frac{1}{2}$				−1		+1			
(1k0)	−3 −4				+2 +1		−4 −3$\frac{1}{2}$		+2 +1$\frac{1}{2}$						+$\frac{1}{2}$ +1

* As defined in the text. The upper figure of each group refers to Δ, the lower to Δ'.

A NOTE ON THE TRANSITION IN ALBITE

As Tuttle and Bowen point out in their accompanying paper, the "low"-"high" albite transformation is very sluggish. In this connection, attention may be called to plate 1, *D*. This photograph is the same as plate 1, *F*, but was exposed for a longer time in order to bring out some diffuse lines between the spots. The pattern of spots is due to the combination of two patterns of the type shown in plate 1, *B*, as a result of albite

twinning. (The b^*-axis coincides for both twin positions, but the a^*-axes do not.) Before heating, the angle between a^* and b^* is greater than 90° (pl. 1, *A*). After heating, this angle is smaller than 90° (pl. 1, *C*). The diffuse lines between the twin spots in plate 1, *D*, indicate states with intermediate values for $a^* \wedge b^*$. Thus the transformation from "low" to "high" albite takes place more or less continuously, $(100) \wedge (010)$ going from 90°30′ to 88°5′.

ACKNOWLEDGMENT.—This work was done in conjunction with a research program financed by the Geophysical Branch of the Office of Naval Research.

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NOTES ON THE FELDSPARS

ON THE RELATION BETWEEN ANORTHITE CONTENT AND γ-INDEX OF NATURAL PLAGIOCLASE[1]

FELIX CHAYES
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Discovery of the close relation between the chemical composition and refractive indices of the plagioclase feldspars was one of the early triumphs of modern mineralogy. Over the entire range of composition this relation is good enough to be extremely useful, but its form has never been determined. That the series is not structurally homogeneous has been established by crystallographers, and departures from continuous variation in the form and orientation of the optical indicatrix have been the subject of elaborate studies by European workers. More recently the existence of high- and low-temperature forms at the albite end of the series has been clearly demonstrated. The literature is reviewed and the experimental work is described elsewhere in this issue. The known high- and low-temperature forms generally differ in index, so that the relation between index and composition must be more complex than was once supposed. Ideally, the whole subject should be thoroughly examined, but this can only be a long-range project. Pending the appearance of someone willing to undertake the task, we have to content ourselves with assembling and reviewing data already published.

Such an approach encounters immense sampling difficulties and can never be more than suggestive, if only for the reason that the data points are not of equal value and tend to be irregularly distributed over the range. It can be shown, however, that index data do indicate—or are, at any rate, compatible with—more complex relationships than the single completely continuous solid solution of classical mineralogy. Without an extensive literature search the intermediate index cannot be used for this purpose, since in many brief notices one cannot determine whether β was measured or computed. The upper and lower indices are available, however, and, though only γ is considered in this note, similar relations appear to hold for α.

Figure 1 is a plot of anorthite content (weight per cent) against the γ-index. Many of the points are averages, particularly at the lower end of the range, but all have been given equal weight in the computations. (Observed anorthite values have been averaged for each recorded index, the indices being read to the third place.) The data points include substantially all the literature values collected by myself and O. F. Tuttle in an intermittent search extending over a few months, but probably not involving a total of more than 10 or 15 hours of library work. The entire group now contains 58 paired analyses and index determinations. Five values were rejected because authors of the original publications apparently regarded them with considerable skepticism. One was discarded because of anomalous sign.

Where correlation is as high as this (the product-moment correlation is 0.997 for the data of fig. 1), the question of which, if either, of the variables is to be regarded as independent or errorless is largely academic. The lines of best fit given by the conventional mean-square calculations are virtually identical. The anorthite content as a function of the γ-index is given by

$$An = 1904.03\,\gamma - 2925.94, \qquad (1)$$

[1] Manuscript received April 17, 1950.

and, with the exception of a few discrepant results, this relation gives an excellent approximation of observed anorthite content over the entire range.

In the region $0 < An \leq 45$, however, the deviations, though mostly small, are curiously systematic. In the first place, all those for which the observed An content is less than 14 are negative, while all those in which observed An exceeds 14 are positive. Second, the size of the deviations decreases almost without break from An_0 to An_{14} and increases almost without break from An_{14} to An_{45}. In order of increasing observed anorthite content the deviations are as follows: -5.8, -3.4, -1.4, -2.6, -1.8, -0.8, $+1.5$, $+1.2$, $+2.3$, $+2.4$, $+3.4$, $+5.4$, $+4.9$. From An_{45} up, the sequence of deviations displays nothing like this regularity in either sign or size.

There is thus a strong suggestion of some systematic departure from equation (1) at the albite end of the series. A line calculated for the region $0 < An < 30$ and extrapolated to An_{45} materially reduces the deviations at the extremes of this range. The calculated equation is

$$An = 2547.31\,\gamma - 3919.27. \qquad (2)$$

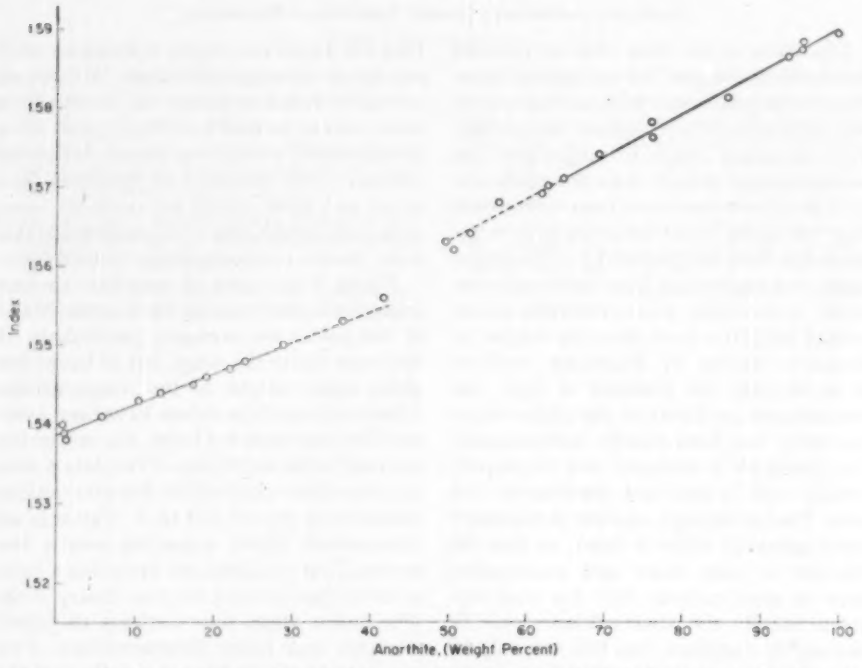

FIG. 1.—Relation between γ-index and an An content of natural plagioclase. (An is the dependent variable of the calculations but is plotted as abscissa to permit comparison with other index-composition diagrams of the plagioclase series.)

It is to be stressed that equations (1) and (2) both describe the data very well; the only question is whether one is better than the other. Because of the peculiarly unsatisfactory sampling technique, it is probably best to refrain from statistical tests. The important matter is whether residual variance,[2]

[2] If variance is used as a measure of variability, then residual variance is that portion of the total variation not accounted for by the regression equa-

a variability unaccounted for by equation (1), is sensibly reduced in this range by equation (2). For the points shown in figure 1, many of which are averages, there is, in fact, an almost fivefold reduction of weighted residual variance if equation (2) is used in place of equation (1). Without weighting, this reduction is more than fourfold; and for the individual values, from which the means have been computed, it is more than seven fold. This would be a highly significant result if the situation permitted formal testing. In any case, purely as a matter of sample description in the specified range, equation (2) is preferable to equation (1). Inspection of figure 1 is enough to show, however, that equation (2) will lead to decidedly erroneous predictions if it is extrapolated much past An_{45}.

If equation (2) is to be substituted for equation (1) in the lower part of the range, a new equation must also be computed for the anorthite end. This new line, calculated for the region $64 \leq An \leq 100$, is

$$An = 1907.28\,\gamma - 2931.51 \qquad (3)$$

and proves to be virtually identical with equation (1).

There is apparently no theoretical requirement that the relation between composition and maximum index be linear, but it is worth noting that departures from linearity in the central part of the range are not of a type that would be substantially reduced by a quadratic term unless the fit at one end were sacrificed. And it is just at the ends of the series that data are most abundant. So far as can be determined from present data, the sample is best described by two straight lines, equation (2) for $An < 45$ and equation (3) for $An > 45$.

Now it happens that the γ-index of synthetic high-temperature albite is considerably less than the γ-intercept value of equation (2), and this suggests a very simple interpretation of the results, namely, that natural plagioclases carrying less than 45 per cent An are usually the low-temperature form, while those with more than 45 per cent An are usually the high-temperature form.[3] Dr. Köhler's (1949) suggestion that volcanic rock-plagioclases are ordinarily high-temperature, while those of plutonic rocks are normally low-temperature, seems at variance with this interpretation; because of the choice of material for index determination, it is not possible to say whether this contradiction is real or apparent. Most of the index determinations for soda-rich compositions have been made on crystals from veins or pegmatites. Nearly all the determinations in the intermediate compositions and many in the lime-rich end of the series are for phenocrysts from lavas. Only in the lime-rich portion of the series, however, are values for truly plutonic plagioclase at all frequent.

Further information over the entire range would be highly desirable and might alter materially not only the sample description but its interpretation as well. Readers will confer a special favor by calling to my attention reliable measurements in the region $30 \leq An \leq 70$.

tion, that is,

$$V_r = \frac{\Sigma(An' - An)^2}{n - 2}$$

where An′ is the observed anorthite content and An is the anorthite content predicted from the regression equation. The value of the ratio V_{r1}/V_{r2} is a measure of the superiority of eq. (2) over eq. (1).

[3] Tuttle and Bowen (companion paper, this issue) record a value of 1.534, whereas γ for An = 0 is 1.539 from eq. (2). Equations (1) and (3) give a γ-value of 1.537, a little closer to the measured value. Of course, if the interpretation offered here is correct, one would hardly expect closer agreement; eq. (3) must be extrapolated a distance equal to its entire range to yield a value for An_0, whereas eq. (1) is based on a mixture of high- and low-temperature feldspars.

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NOTE ON "PSEUDO-MONOCLINIC" PLAGIOCLASE

CHRISTOFFER OFTEDAHL
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In a recent paper (1948) the author has described the feldspars occurring in the igneous rocks of the Oslo region, Norway. Later Köhler (1949) and Buerger (1948) advanced some new views upon the problem of the high-temperature plagioclases. These viewpoints are well illustrated by plagioclases of the Oslo region, and therefore a few of the characteristics of these feldspars will be pointed out below.

The uncertainty about plagioclase optics was definitely cleared up when Köhler (1941) found that there are two series of plagioclases with different optics: (1) the high-temperature plagioclases, formed in volcanic rocks, and (2) the low-temperature plagioclases, formed in plutonic and metamorphic rocks. On the basis of optical study by the present author (1948, p. 14) it is not certain at present whether these plagioclase varieties display polymorphism.

Köhler (1949, p. 598) has assumed that primary plagioclase which crystallized in melts was monoclinic, or nearly so, and that the finely laminated albite twinning in such plagioclases is induced in the plagioclase by the change from monoclinic to triclinic symmetry. Moreover, Buerger (1948, p. 120) mentioned parenthetically that plagioclase may have a monoclinic high-temperature form.

The Oslo plagioclases give excellent examples of how plagioclases may have crystallized as monoclinic crystals under high temperatures and low pressures, with later transformation to triclinic symmetry involving the least possible change of the monoclinic lattice.

The feldspar phenocrysts of the rhomb porphyries were assumed by W. C. Brögger to be lime-bearing soda orthoclases, locally soda microclines. This statement is frequent in present-day textbooks of petrography and mineralogy. The feldspars, however, are actually plagioclase, although they are optically monoclinic, owing to very fine albite twinning. The lamellae may show up only when the twin plane is exactly parallel to the microscope axis. Therefore a universal stage is necessary for the investigation of such feldspars.

The oligoclases of deep-seated rocks show the same phenomena, although with slightly broader lamellae in general. Thus traces of twinning may be seen in a few plagioclase individuals in rock sections.

Basic igneous rocks contain plagioclases with broad lamellae, and the width decreases with decreasing An content. The lamellae "disappear" at a composition An_{40} in volcanic rocks, whereas the plagioclases in plutonic rocks become "pseudo-monoclinic" at a composition An_{30}.

This plagioclase phenomenon also occurs in the alkali feldspar group. The alkali feldspars of monzonitic and of some syenitic rocks are soda-rich cryptoperthites or cryptoperthites transitional to microperthites. Most cryptoperthites are monoclinic, though they are composed of submicroscopic lamellae of orthoclase and (triclinic) albite. Weissenberg photographs showed that this plagioclase is twinned according to the albite law. The phenomenon obviously occurs in all moonstones.

Thus these Oslo plagioclases may be considered to have formed from monoclinic crystals with as little structural disarrangement as possible during the cooling.

The plagioclases of volcanic rocks and of at least some intrusive rocks show high-temperature optics (Oftedahl, 1944, p. 75). More interesting is the fact not only that optical data suggest the existence of high-temperature optics in some plutonic plagioclases but that some X-ray data suggest the same optics in the plagioclase phase of cryptoperthites.

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REVIEWS

Geology: Principles and Processes. By WILLIAM H. EMMONS, GEORGE A. THIEL, CLINTON R. STAUFFER, and IRA S. ALLISON. 3d ed. New York: McGraw-Hill Book Co., Inc., 1949. Pp. x+502; figs. 488. $4.75.

Laurence M. Gould, in a review of the second edition of this text which appeared in the *Journal of Geology* in 1939, described it as "a book which the student will find readable and the teacher find teachable." This is equally true of the third edition, which contains many aerial photographs that add to the value of the book.

However, several misprints occur. On the cover is imprinted *Principles and Practices* instead of *Principles and Processes*. In the Preface the statement is made that a new unit, chapter v, has been added. It turns out to be chapter vi. On page 95 there is a reference to the petrified logs and forests of Arizona shown in figure 84. Figure 84 is a photograph of petrified tree stumps from Yellowstone National Park. On page 274 reference is made to a beach shown in figure 276; that figure is a print of a natural bridge.

On page 269, figure 267, labeling would aid the beginning student in differentiating between a diatom, a radiolarian, and a sponge spicule. On page 252 is a statement referring to the continental shelf: "It slopes gradually from the shore to about the 100-fathom line, where the bottom begins to descend more abruptly into abysmal depths." According to investigations by Shepard (1939), "The use of the 100-fathom line as an outer limit is unfortunate in that steepening beyond the shoal platforms is initiated at an average depth of about 65 fathoms."

DUNCAN STEWART, JR.

The Amadeus William Grabau Memorial Volume. (Geological Society of China Bulletin, vol. 27.) 1947. Pp. 398.

Dr. Grabau was one of the most remarkable geologists of modern times. In 1920, at the age of fifty and after a successful career as teacher and research worker in this country, he went to China as professor of paleontology in the National University of Peking and as chief paleontologist of the Geological Survey of China. There he met an opportunity such as comes to few men and became the outstanding figure in the development of modern geology in his adopted country. He trained and supervised the work of a generation of stratigraphers and paleontologists, whom he imbued with his enthusiasm and respect for high standards in scientific work. He also published results of important personal research and several volumes of compiled geological data and theoretical interpretations that are well known to geologists in all lands. In influence, fame, and the respect that he won from all, he can only be compared with some of the ancient Chinese heroes, and, if he had lived in an earlier age, he undoubtedly would have been deified.

Dr. Grabau died in 1946, six months after the liberation of China, and this volume was prepared as an inadequate but most sincere memorial to an undoubtedly great man. It begins with three memorial and biographical papers, including a complete bibliography. About half the succeeding scientific papers are devoted to the fields of Grabau's particular interests, stratigraphy and related subjects (6), and paleontology (6), but also broadly cover other geologic fields: structure (3), sedimentation (2), petrography (2), economic (2), and one each on physiography, glaciology, and crystallography. The twenty-three Chinese contributions include many of the best-known geologists of that country, and among the seven foreign contributors are several who have long been interested in the geology of China.

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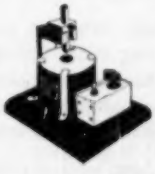

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